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Impact of wastewater reuse on cobalt status in Egyptian environment

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Abstract: Cobalt is used in the manufacture of alloys, catalysts in the petroleum industry, catalytic converters, and paint pigments. Thus the potential for Co releases into the environment is highly increased. Use of waste sludges and sewage effluent to fertilize and irrigate soils has also increased soil Co concentrations. Total cobalt contents of alluvial delta soil of Egypt show considerable variation ranging from 13.1 to 64.7 ppm. The impact of either wastewater irrigation or industrial activities on soil total Co was obvious due to accumulation of organic matter and solid waste in the surface soil samples. Food crops and vegetables should not be grown on soil highly contaminated by Co. It is noteworthy that the delayed neutron activation analysis(DNAA) technique could be used successfully for total Co determination due to its high sensitivity. It is quit clearly that dust samples of Cairo City contains higher Co level, as compared to Suez Canal Region(Ismailia, Port Said and El-Suez cities). The high values in Cairo City may be due to the existence of industries around the city and the intensive traffic. To minimize Co environmental hazards, waste effluents should be treated on site. Thus, levels of potentially toxic Co needs to be continuously monitored and should be removed during several treatment processes before the disposal of these wastes.

Keywords: cobalt; environment; Egypt

Introduction

In nature Co occurs in two oxidation states, Co^{2+} and Co^{3+}, and formation of the complex anion $Co(OH)_3^-$ is also possible. During weathering Co is relatively mobile in oxidizing acid environment, but due to a high sorption by Fe and Mn oxides, as well as by clay minerals, this metal does not migrate in a soluble phase. Concentration of Co in soils is inherited from parent materials. Soils over mafic rocks and soils derived from clay deposits contain the highest amounts of this metal. Alloway(Alloway, 1990) mentioned that naturally high Co contents are usually observed in soils over serpentine rock and ore deposits. Significant sources of Co pollution are related to nonferrous metal smelters whereas coal and other fuel combustion are of considerably less importance, however, roadside soils and street dusts known to be enriched in Co.

The primary interest in Co as constituent of soils lies in its essential roles in ruminant animals and microorganisms, and because of deficiencies rather than excesses. Interest in Co in plant biology stems particularly from its essential role in biological N-fixation. However, large quantities of Co are added to soils via its widespread use in many industrial activities. Cobalt is used in the manufacture of alloys(Purves, 1977), catalysts in the petroleum industry, catalytic converters, and paint pigments(Hunter, 1975). Use of waste sludges and sewage effluent to fertilize and irrigate soils has also increased soil Co concentrations(Abdel-Sabour, 2000).

Aubert and Pinta(Aubert, 1977) mentioned that total Co contents of soils vary widely from 0.05 to 300 ppm with an average content in the range of 10.0—15.0 ppm. Kobata-Pendias and Pendias(Kobata-Pendias, 1984) noticed higher Co content in the surface layer of soils in arid and semi arid regions. The contents of Co in soils vary mainly in relation to the parent materials from which they were derived, even though there are also differences with depth in the soil profile and between soil types derived from common parent materials due to pedological processes. Within a given soil profile, Co is generally concentrated in those horizons rich in organic materials and clays.

When a high Co level is readily available, in polluted soil in particular, it can seriously affect plant growth and metabolic function. Data is scarce concerning Co pollution and there are no reports of Co toxicity to animals or human attributed to consumption of natural feed and / or foodstuffs. However, under the influence of man induced pollution, the excess of Co in soils and plants may be a health risk.

1 Total cobalt in the soil

The normal Co content of surface soils usually ranges from 1.0 to 40.0 ppm, with the highest frequency in the range of

3.0 to 15.0 ppm. The grand mean Co concentration for worldwide soils is 8.5 ppm and for the soils of the US is 8.2 ppm. Usually higher Co contents of surface soils are observed for arid and semiarid regions(Kobata-Pendias, 1984).

In Egypt, Rashad *et al*.(Rashad, 1995) found that the normal level of Co in the alluvial soils of Nile Delta ranged between 3.7 and 5.5 ppm with an average of 4.7 ppm for total content and from 0.04 to 0.11 ppm with an average of 0.07 ppm for available from El-Aggory *et al*.(El-Aggory, 1993) investigated total and available Co in 25 surface soil samples representing different geographical sites in the Nile valley. He indicated that Co levels are well within the ranges for Co levels (Table 1) in normal soils given(Kobata-Pendias, 1992).

Table 1 Total and available Co in different soil types(ppm)

Soil	Alluvial heavy texture			Alluvial light texture			Calcareous soils		
	Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
Total	30.1	37.70	32.20	17.4	33.80	24.90	10.3	20.7	15.50
Available	0.02	0.28	0.16	0.12	0.62	0.43	0.00	0.13	0.09

1.1 A study case

The aim of this study was to evaluate total Co content in alluvial soils of Delta in Egypt using the neutron activation analysis technique(NAA).

1.1.1 Soil sampling

Twenty-five surface(0—20 cm) soil samples were collected from different locations in Egypt representing non-polluted, moderately and highly polluted soils. The non-polluted soil samples were collected from different sites not affected by any known sources of Co pollution. The moderately and highly polluted soils samples were collected from soils subjected to prolonged irrigation with either polluted industrial wastewater or samples collected from industrial sites e.g. metaliological workshops. The samples were air-dried, crushed pass through a 2.0 mm sieve then the following analysis were conducted. Mechanical analysis was carried out by the pipette method. Soil pH and organic matter content was determined by standard method as described by Jackson(Jackson, 1969). Calcium carbonate content was determined volumetrically using Collin's calcimeter. One gram of each soil samples was wet digested and total Co was determined using atomic absorption spectroscopy (AAS) according to Jackson(Jackson, 1969).

1.1.2 NAA procedure

Soil samples were sieved through 800 mesh sieve to obtain fine homogenous samples. About 0.1 g of each sample was packed in pure aluminum foil and prepared for irradiation along with a capsule of standard reference materials(standard sample soil-7; IAEA-SL7) for the purpose of relative calculation and for analytical quality assurance testing. An empty aluminum foil of known weight was irradiated at the same conditions, for identifying and subtracting the background γ-ray lines due to the aluminum envelopes. Then all samples were sealed well in aluminum cans. The irradiation time was 48 h at the Nuclear Research Center. First Research Egyptian Reactor 2MW(ET-RR-1), the neutron flux was 4.4×10^{12} n/(s·cm^2). After 72 h (cooling time) the samples were radio-assayed for γ-ray spectra using high resolution hyper pure Ge detector connected to multi-channel analyzer through a suitable electronic system including the PCA computer program. The HP-Ge detector was model No. 15190 Coaxial type with a crystal of 76.11 cm^3 and full width at half maximum(FWHM) of 1.72 KeV at 1332.5 KeV γ-ray line of Co^{60} with an efficiency of 15% and a peak to Compton ratio of 55. The detector and the attached electronic circuits in conjunction with the computerized multi-channel analyzer were used for measuring the induced radioactivity. Energy and efficiency calibration curves up to 3 MeV at the experimental conditions were carried out using the multi-γ-ray standard source MCS-4 (NMG-MAG, 1994).

1.1.3 Measurements

The samples were positioned individually at about 10 cm in front the detector and the accumulating time for two hours to assure good statistics. Cobalt-60 concentration was calculated from measurements of γ-activity induced by ^{59}Co(n, γ) ^{60}Co reactions. The two prominent gamma ray lines at 1173.2 and 1332.5 KeV were efficiently used for ^{60}Co determination. Cobalt concentrations in soil samples were determined using the equation of comparative standard method(Ehman, 1991).

1.2 Results and discussion

To satisfy the accuracy and precision of the detection system, the standard reference material(soil-7) was analyzed. The well-resolved Co^{60} and Cs^{134} lines clearly appeared and had been compared with the certified values as shown in Table 2. The data of the present work showed a good agreement with the certified values.

Table 2 Analysis of standard reference material IEAE-soil 7

Elements	Present work, ppm	Certified values, ppm	Ratio
Co^{60}	8.07 ± 0.4	8.9	0.906
Cs^{134}	5.14 ± 0.25	5.4	0.951

Table 3 shows some selected soil characteristics along with total Co concentrations determined by NAA technique. Co content in non-polluted soil samples ranged between 13.12 to 23.20 ppm with an average of 16.9 ± 4.38 ppm. These values are relatively higher than reported by Rashad *et al*.(Rashad, 1995). They reported that normal levels of Co in the alluvial soils of Nile Delta ranged between 3.7 and 5.5 ppm with an average of 4.7 ppm.

Cobalt content in moderately polluted soils ranged between 26.5 to 30.00 ppm with an average of 28.3 ± 1.3 ppm. The highest Co levels ranged from 36 to 64.69 ppm with an average of 51.9 ± 9.5; were observed in soil samples collected from, either highly polluted agricultural soils due to prolonged irrigation with industrial wastewater(i.e. soils of Mostord, Shubra El-Kima and Balaks Qalubia governorate), or surface soil samples from industrial sites as shown in Table 3.

Table 3 Cobalt concentrations in alluvial soil samples collected from different Delta sites (non-polluted and polluted) as determined by NAA and AAS techniques

Location	Soil properties					Total Co, ppm	
	PH	O.M, %	$CaCO_3$, %	Clay, %	Silt, %	NAA	AAS
Non polluted sites							
Bahtim #1	7.8	2.40	2.15	45.80	31.50	13.20	12.01
Bahtim #2	8.1	2.15	1.80	43.80	35.50	14.10	10.49
Bahtim #3	8.2	1.15	0.85	39.90	41.20	13.12	8.79
Kaluob #1	8.1	1.25	0.72	43.55	40.85	13.80	11.94
Kaluob #2	8.1	1.51	3.34	34.78	32.00	14.00	9.54
Kaluob #3	8.2	1.30	3.06	33.03	35.50	15.00	13.80
El-Qanater A	8.2	0.59	3.15	47.03	41.20	14.00	8.99
El-Qanater B	8.3	0.71	2.50	54.13	40.85	15.00	10.75
Qena	7.9	1.44	6.44	30.90	25.55	20.60	18.78
Giza	7.9	2.80	3.87	26.65	28.05	25.20	24.01
Tanta	7.7	3.37	3.40	38.50	19.15	21.30	18.74
Mansora	7.7	1.32	3.07	3.04	42.06	23.20	19.89
Moderatly pollutedsites							
Idfu	7.8	1.74	8.85	31.75	26.25	26.50	24.55
Zagazig	8.0	2.65	3.24	30.05	31.15	27.90	23.74
Minya	8.5	2.28	3.71	10.75	39.15	28.60	25.63
Damietta	7.7	3.00	3.26	43.00	20.95	28.50	26.61
Disuq	7.7	7.73	3.56	36.60	24.70	30.00	28.50
Highly polluted sites							
Mostord (A)	8.0	2.70	4.20	55.60	23.50	62.50	58.31
Mostord (B)	8.1	2.40	5.20	62.20	29.30	49.50	42.20
Shubra El-Kima	7.2	2.26	2.70	33.15	33.73	47.00	43.56
Balaks	7.5	1.76	1.74	34.70	31.50	36.00	34.50
Industrial sites #1	7.9	5.94	3.94	35.20	31.80	44.99	42.12
Industrial sites #2	8.2	6.33	4.35	42.50	35.60	64.69	52.05
Industrial sites #3	7.5	3.70	3.70	39.90	42.80	56.83	49.91
Industrial sites #4	7.5	3.82	3.82	34.20	33.90	53.83	49.35

Concerning contaminated Egyptian alluvial soils little work was done on cobalt. El-Leithi(El-Leithi, 1986) studied the effect of industrial activities on the soils of Nile Delta. He found that total content of Co in these soils ranged between 11.2

and 36.1 ppm with an average of 23.7 ppm. El-Gamal(El-Gamal, 1980) found that total Co ranged from 30.36 to 41.40 ppm with an average of about 36.5 ppm in El-Gabal El-Asfar sandy soils irrigated with sewage sludge for several years. The surface layers containing higher amounts of Co than the subsurface ones. Similar results were obtained by Rashad and Khalil (Rashad, 1996; Khalil, 1980). Fig. 1 shows Co mean values of the tested non-polluted, moderately and highly polluted soils.

The comparison between total-Co values determined by either NAA or AAS methods showed a significant difference, particularly at soils with low Co-levels such as the non-polluted samples. However, a very strong linear relation was observed as shown in Fig.2. The obtained regression equation with significant R value (0.982) suggested that Co determined by NAA method always was higher than the relevant values determined by AAS method. Caplan *et al*. (Caplan, 1987) indicated that Co can be determined adequately by NAA at ambient levels if compared by AAS method.

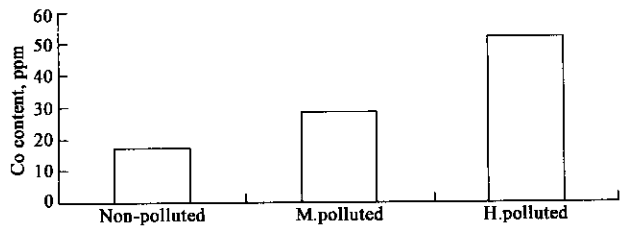

Fig. 1 Mean values of total Co in soils as affected by pollution

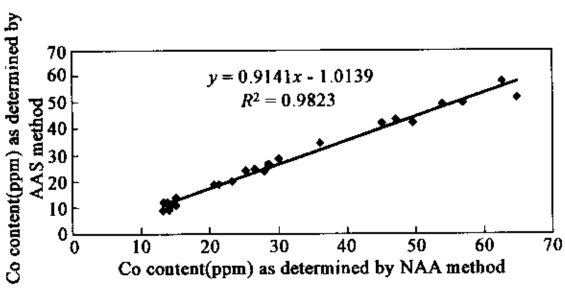

Fig. 2 Comparison between Co values as determined by two analytical methods(NAA and AAS)

2 Cobalt in plant

2.1 Field crops content

An area suffers from several environmental problems such as industrial wastewater and sewage effluent disposal. Some of the agricultural fields are irrigated by wastewater directly without any treatment. The analysis of water samples showed that El-Shaboura canal and Shebin El-Qanater collector drain exhibited the highest levels of Cd, Co, Pb and Ni. While Mostord collector drain has the highest levels of Zn and Cu (Abdel-Sabour, 1998). The area was divided to six sectors according to the source of irrigation water and the probability of pollution with organic and inorganic wastes (Fig. 3). Sectors A and B represent non-polluted soil and are taken as control. Sector C represents soils subjected to irrigation with heavily polluted wastewater from Shebin El-Qanater collector drain (receives wastewater from plastic, staining and fabric factories). Sector D represents soils subjected to irrigation with polluted water (domestic dumping). Sector E represents soils subjected to irrigation with combination of El-Shaboura canal water and either Mostorod collector wastewater or Shebin El-Qanter collector wastewater drain. Sector F represents soils subjected to irrigation from El-Boulaqeyah canal water and Laza agricultural drain wastewater. Sector G represent soils subjected to irrigation with heavily polluted wastewater of Mostorod collector drain, which receives wastewater from steel, staining, paper, and detergents factories. Abdel-Sabour and Rabie (Abdel-Sabour, 2000a) investigated Co content in filed crops grown on alluvial Nile Delta soil (either normal or contaminated ones due to irrigation with untreated industrial wastewater). They indicated that the investigated crops accumulated high contents above the normal range(0.53—1.63 ppm) according to Aboulroos *et al*. (Aboulroos, 1996) especially in case of sorghum. The highest Co content was observed for plant samples collected from contaminated soils in the following order C > E > F.

2.2 Cobalt in vegetable

In another study on the same area Abdel-Sabour and Rabie (Abdel-Sabour, 2000b) investigated Co-content in ten vegetables plant namely, abelmashus, carden, onion, jews mallow, spinach, radish, celery, chard, rocket and lettuce grown on either normal or contaminated soil of the same origin. It is clear from the study that vegetables grown on contaminated soil C and D showed the highest accumulation of Co especially the leafy plant such as lettuce, spinach, carden, rocket and radish.

Presented data (Table 5) shows that vegetables grown on soils of groups C, D and G have the highest content of Co in their tissues. Most of the investigated plants accumulated high amounts of Co, which exceeds very much the normal range. Rocket recorded the highest content of Co (61.3 μg/g) followed by celery (39.7 μg/g), spinach (32.3 μg/g), jews mallow (32.1 μg/g), onion (31.5 μg/g), lettuce (29.4 μg/g), chard (28.8 μg/g) and carden (25.7 μg/g). Though radish and abelmashus recorded the lowest contents of Co (11.6 and 13.7 μg/g, respectively), but these levels exceeds the normal range as reported by Bowen (Bowen, 1979). He reported that the concentration of Co in edible vegetables ranged between 0.01 and 4.60 with an average of 2.31 ppm.

Table 4 Cobalt content (μg/g) in selected grain crops as affected by different contaminated irrigation waters in tested area

Soil	Soil	Barley	Wheat	Maize	Rice	Sorghum
Normal delta	A	ND	ND	ND	ND	ND
soil (control)	B	0.60	0.70	0.75	0.5	1.6
Contaminated	C	5.80	6.80	8.35	3.1	16.7
soil	E	2.80	2.00	4.20	1.5	12.8
	F	1.90	1.80	1.00	1.8	10.8
Mean of (C, E, F)		3.50	3.53	4.52	1.5	10.8

Table 5 Heavy metals concentration (μg/g) in tested vegetables plant grown on contaminated soils in tested area

Site	A*	B**	C	D	E	G
Abelmashus	ND	1.6	8.5	5.8	3.2	
Carden	ND	2.7	15	11	6	
Onion	ND	0.2	1.1	0.8	0.5	
Jews mallow	ND	1.2	6.5	4.7	3.1	
Spinach	ND	2.5	13.6	9.8	3.2	
Radish	ND	2.1	11.8	8.2	5.1	10.8
Celery	ND	1.2	10.1	6.2	3.8	8.8
Chard	ND	1.1	6.6	4.4	2.6	6.5
Rocket	ND	3.7	19.9	11.2	6.5	18.7
Lettuce	ND	3.8	22.8	15.6	9.4	18.6

A*: normal delta soil; B**: control soil in the tested area

3 Cobalt in water

3.1 Fresh water

In a study (Table 6) conducted by General Authority of Industrialization (personal communication) dealing with effluent discharging points of Governmental Industrial Units on the Nile, results reveal that Co levels ranged between 10 to 74 μg/L (the highest values observed in polluted spots) along the river. Lower Co levels (between 10—20 μg/L) were reported by El-Falaky *et al*. (El-Falaky, 1988). Recently Abdel-Sabour and Rabie (unpublished data) reported that water samples collected from River Nile Northern Cairo and from Ismailia Canal averaged as low as 7.7 ± 5.5 μg/L. Abdulh and Royle (Abdulh, 1974) reported that average of Co level in fresh water may range between 0.2—27 μg/L.

Table 6 Cobalt levels (μg/L) along River Nile at different industrial discharging points

Site	Water body	Co, μg/L
Helwan industrial complex	Nile	74
Sugar and distillation factory	Nile	39
Sugar refienery	Nile	42
Upper pump	Damietta	16
Upper Serw pump	Damietta	12
Rahawi drain	Rosetta	22
Sabal drain	Rosetta	19
Total drain	Rosetta	10
Sahand soda factory	Rosetta	65
Pesticide factory	Rosetta	36
Malla factory	Rosetta	53

3.2 Drainage water

Water quality of different Egyptian drains depends on the nature and amounts of the wastewater load discharged to the drain. The flow-rate of both current stream and discharged waste effluent has a crucial role on drains water quality. The Soil and Water Research Institute surveyed mid-delta drainage waters in 1990. The Co content was reported to be in the range of 6 to 27 μg/L with an average of 26 μg/L.

3.3 Cobalt in wastewater

The above mentioned studied area (Shoubra El-Khima, Bahteem and Mostorod which lies in the industrial area north of Greater Cairo) includes several different industrial activities such as plastic, steel, fabric, staining, paper oil & cleaners industries which discharge their liquid wastes into different water bodies (Abdel-Sabour, 1998). Also, there are various small factories and workshops, which discharge their wastewater into the sewer system or transport their solid and liquid wastes by trucks to the previous water bodies. Another pollution sources are the sewage effluents and domestic wastes that are discharged to water bodies through many pipes at different sites(Abdel-Sabour, 1998). Water samples were collected seasonally from different waterways found in the area, domestic and/or industrial liquid wastes from 12 discharge tubes of different factories(as a point source of pollution). Three drains namely; Shebin El-Qanater and Mostorod collector drains which are being used for the disposal of all types of wastes(liquid and solid industrial wastes and sewage effluents), and agricultural laza drain. Chemical characteristics of different water samples and its heavy metals content were determined using ion coupled plasma technique(ICP). Results indicated that industrial and domestic wastewater samples contain several toxic levels of tested heavy metals(Cd, Co, Pb and Ni), which have a serious impact on surface waterways in the area. Shebin El-Qanater collector drain samples exhibited the highest levels of Cd, Co, Pb and Ni compared to other tested water bodies. Industrial effluent samples collected from Cairo Company for Fabric industry had the highest amounts of total Zn Cu, Cd, Co and Pb. Generally, it is necessary to minimize the risk of these pollutants by regulating the industrial wastewater disposals.

Data in Table 7 show Co concentrations (μg/L) and the amount of wastewater discharged annually from investigated factories as well as their content of Co (kg/a). The highest Co concentration was recorded in the wastewater of Cairo Staining Company (128 μg/L). The lowest values were observed in the Egyptian Company for Starch (4 μg/L). Concerning the amounts of Co discharged annually, Cairo Fabric recorded the highest values (471 kg/a) followed by Egyptian Company for Paper (270 kg/a). The lowest Co values were recorded in the Electric Company wastewater(1.2 kg/a).

Table 7 Mean values for total Co concentrations (μg/L) in the liquid wastes of some industrial factories and domestic wastes during the period 1993 – 1995

Factory	Co, μg/L	Annual load	
		Amount of wastewater, million m^3/a	Co(kg/a) in liquid wastes during the period (1993 – 1995)
Ahleya Blastic Company	9	4.40	40
Electric Company	12	0.10	1.2
Abdel-Gabbar Staining Co.	24	0.12	2.9
Cairo Co. for Staining	128	0.99	127
Cairo Co. for Fabric	92	5.12	471
Delta Co. for Steel	20	4.30	86
El-Salam Staining Co.	69	0.18	12
Nile Co. for Oil and Cleaners	57	2.20	125
El-Nogoum Staining Co.	81	0.16	12.9
Egypt Co. of Starch	4	2.60	9.2
Politex Staining Co.	89	0.85	75.7
Egypt Co. for Paper	367	3.60	270
Sewage effluents	252	0.37	14.2
Domestic wastes	148	0.075	1.4

Fig.3 shows the average concentrations of dissolved Co as well as Co-contents in suspended matter in contaminated water bodies at the studied area. The wastewater of Shebin El-Qanater and Mostorod collectors recorded the highest levels of dissolved and suspended-Co during all seasons while Laza drain recorded the lowest values.

4 Atmospheric fall-out of Co

Ali and Elsikhry(Ali, 1993) investigated the total amounts of cobalt in the aerial dust settled on surface leaves of *Ficus nitida* grown in Cairo Governorate and Suez Canal Region. From their data, it is quit clearly that dust samples of Cairo City contains higher Co level, as compared to Suez Canal Region(Fig.4). The high values in Cairo City may be due to the existence of some industries around the city and the intensive traffic.

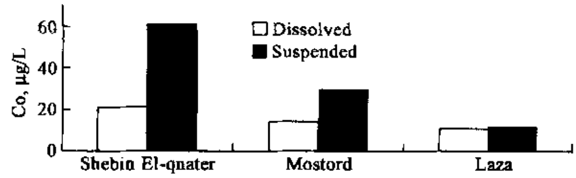

Fig. 3 Mean of dissolved and suspended Co in different water samples

El-Ghandour *et al*.(El-Ghandour, 1982) investigated Co levels in dust deposits in various sites in Helwan industrial area (24 km south-east Cairo). They indicated that Co was found in trace amounts ranging between 0.01% and 0.03% of the total annual dust fall. Their data showed clearly increasing Co levels in night dust fall samples. This can be attributed mainly to the

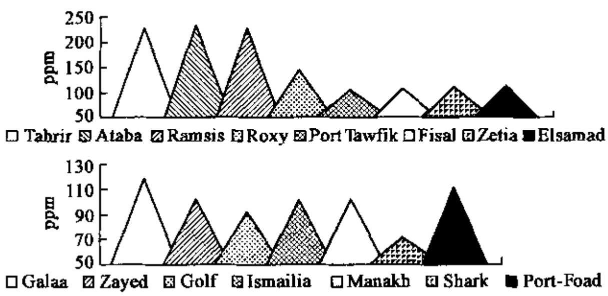

Fig. 4 The total amounts of cobalt in the aerial dust settled on surface leaves of Ficus nitida grown in Cairo Governorate and Suez Canal Region

nature of the industrially emitted metal particles, being of small size and should have a long residence time in the air. This process depends greatly on the local climate of the study area. For example, rare rainfall and *relatively high temperature through most of the* year characterize Cairo. Consequently, the daytime heating has the ability to produce intense mixing of air mass which results in an increase of the fine particles in the atmosphere.

5 Conclusions

Generally, results reveal that industrial waste contain toxic amount of Co with relatively high levels and may have a serious impact on the surrounding environment, as they do not degrade and when accumulate they cause adverse effect on the environment. To minimize the environmental hazards, waste effluents should be treated on sites. Thus, levels of potentially toxic Co needs to be continuously monitored and should be removed during several treatment processes before the disposal of these wastes.

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