



Application of pollution indices for evaluation of heavy metals in soil close to phosphate fertilizer plant, Assiut, Egypt

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ABSTRACT

Pollution indices were used to investigate the levels of heavy metal concentrations in a polluted area under the effect of fertilizer plant emissions at Manqabad, Assiut. Huge amounts of dusts, fumes and gases containing heavy metals which are released from the industrial processing activities, precipitated in the neighbouring area by the effect of meteorological factors causing soil pollution. Soil samples were taken from the area downwind of the factory and analyzed to detect concentration of heavy metals in the study area. Results of soil analysis showed that the area has been harmfully affected by the heavy metals: Cd, Cu, Pb and Zn and their average concentrations are 0.3 mg/kg, 57.0 mg/kg, 94.7 mg/kg and 80.7 mg/kg respectively. These values exceeded the concentrations in soil background (0.013 mg/kg for Cd, 9.62 mg/kg for Cu, 5.17 mg/kg for Pb and 11.56 mg/kg for Zn) obtained from unpolluted area far from industrial activities and also exceeded the international standards for soil pollution stated by WHO. Pollution indices were developed to evaluate the levels of soil pollution with heavy metals. Index of geo-accumulation (Igeo), Enrichment Factor (EF), Pollution Index (PI), and Load Pollution Index (LPI), were estimated. The calculated results of contamination degree showed that the investigated area lies under the class of high pollution. In final conclusion, the investigated area is highly affected by emissions from the fertilizer plant. So control measures should be applied to the fertilizer plant and the polluted soil area adjacent to the factory should be remediate.

Keywords: Heavy Metal, Fertilizer Plant, Soil Pollution, ICP-MS, Pollution Index, Enrichment factor, Geo-accumulation index, Contamination degree, Assiut

INTRODUCTION

In the last two decades, many researches and studies concentrated on the contribution of heavy metals in the environment and their hazardous effects in the environmental pollution. Heavy metals are released in the environment from industrial activities, agriculture, mining and metallurgical processes and vehicular emission. (Yammine *et al.*, 2010) reported that heavy metals contamination in the biosphere by human activities has become an important process in the geochemical cycling of these metals. Scientific research has proved that stationary and mobile sources of industrial areas caused a release of large quantities of

heavy metals into the atmosphere, soil and plant which exceed the natural levels. (Ruhling *et al.*, 1998) evaluated that pollution of heavy metals is a problem of concern due to their undegradable conditions and toxic to living organisms if they exceeded certain limits.

Heavy metal toxicity has an inhibitory effect on plants growth, enzymatic activity, stomata functions, photosynthesis activity, microbial activity and accumulation of other nutrient elements and also damages the root system (Johansson *et al.*, 1998). On the other hand, beside the soil is a medium for plant growth or pool to dispose of undesirable materials, it serves as a transmitter of many pollutants to surface water, groundwater,

atmosphere and food (Hani *et al.*, 2010). As a result, soil pollution may cause human health risk through its effects on the hygiene quality of food and drinking water. Also soil pollution affects air quality through enrichment of trace metal content in air borne particles originating from soil.

Atmospheric emissions from industrial complexes are considered as the main source of the environmental pollution. The particulates associated with heavy metals which are emitted from phosphate fertilizer manufacturing may contribute to a large scale in deteriorations of the environment. These emissions travel along vast areas by the effect of the meteorological factors and accumulated in soil, plant, animal and may reach the food chain (Mohamed, 2006).

In this study some heavy metals such as: Cadmium, Lead, Copper and Zinc associated with particle emissions from the phosphate fertilizer manufacturing plant have been determined using Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS). Also

physicochemical properties of heavy metal in soil were detected. To understand the pattern of heavy metal distribution, pollution indices such as: pollution index, geo-accumulation index, enrichment factor and degree of contamination were used as indicators of pollution (Chen *et al.*, 2005).

The main objectives of the present study were to assess heavy metal contamination released from a phosphate fertilizer plant in Assiut city in Upper-Egypt. Pollution indices have been used to interpret heavy metal in the vicinity contamination in soil of the area of the fertilizer plants.

MATERIALS AND METHODS

Study Area

The super phosphate fertilizer factory is located at 9 km north of Assiut city, (27° N and 31° E) Upper - Egypt. It lies between the Nile river (East) and Ibrahemia canal (West) as illustrated in Fig. 1.

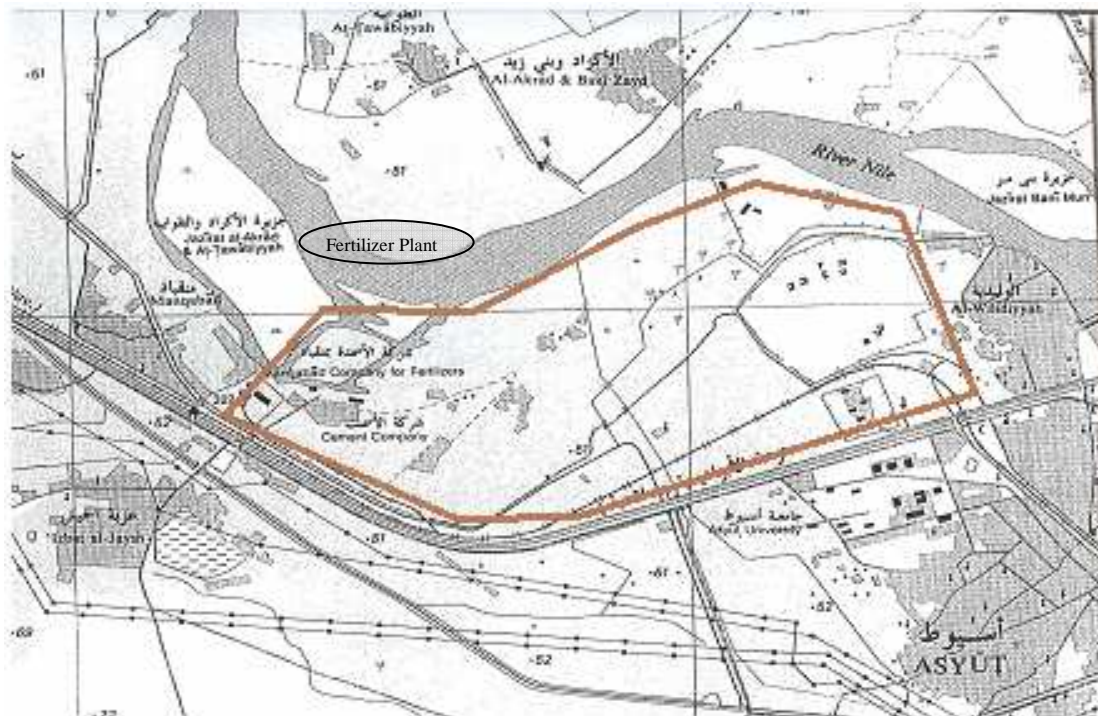


Fig. (1): Location of Phosphate Fertilizer Plant

The area around the factory was cultivated with some main crops such as wheat, faba bean and clover in winter and sorghum, maize and cotton in summer. In addition, some fruit orchards like grapes, banana, jawava and figs are found in the area. The area under investigation is an agricultural land of 100 km² and inhabited with 20,000 residents in five scattering communities.

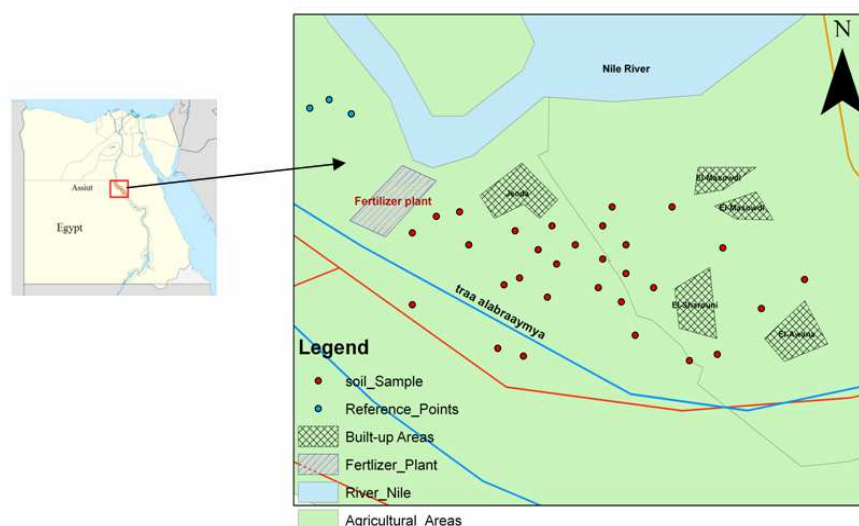


Fig. 2: Location of the investigation area and sites of soil samples

Sampling Sites

Fig. 2 represents the location of study area and the site of soil samples downwind the fertilizer plant as well as locations of control soil samples. A total of 31 topsoil samples were collected from the investigated area. From each location of soil, five sub-samples were collected from the top layer of the soil (15 cm beneath the surface layer). After removing the debris, rock pieces and physical contaminants, the composite samples were placed in polyethylene bags and were brought to the analysis laboratory. Background concentrations of heavy metals in soil samples were collected from upwind distance far from phosphate fertilizer plant to about 3 km, not affected by anthropogenic activities. Location coordinates for all sampled sites were determined using Global Positioning System (GPS) where data are to be used by Geographical Information System (GIS) for data processing.

Soil Samples Preparation and Chemical Analysis

The samples were brought to the analysis laboratory where they were air dried and mixed thoroughly to obtain the representative samples. Soon after drying small debris and other objects were hand picked up and the samples were grinded in a mortar to break up the aggregates or lumps, taking care not to break actual soil particles. Soil samples were then passed through a 2 mm sieve in order to collect the fraction of coarse

particles. Since trace metals are often found mainly in clay and silt fractions of soil, the size fraction $< 63 \mu\text{m}$ is most commonly the recommended size. The granulometric fraction was added with the dispersing agent and after shaking the sand fraction was separated from the clay and silt with $< 63 \mu\text{m}$ sieve, and samples were analyzed to measure the concentration of the heavy metals Cd, Pb, Cu, and Zn. The clay and silt fraction were digested by acids to get the solution by taking 5g of sample into a 300 ml polypropylene wide-mouthed jar and distilled water was added to make a total 200 ml. Then it was acidified with 10 ml HF, 5ml HClO₄, 2.5ml HCl and 2.5ml HNO₃ in order to complete digestion of soil. This jar was shaken on an orbital shaker for 16 hours at 200-220 rpm before being filtered through Whatman filter paper (No. 42) into acid washed bottles. The solution was stored and heavy metal contents were analyzed by Inductive Coupled Plasma Mass Spectroscopy (ICP-MS). Soil pH was determined using digital pH meter according to the method described by **Bates (1954)**. Soil electric conductivity was determined using conductivity meter according to the method outlined by **Godson et al. (2005)**. Organic Carbon and organic matter were determined according to the method outlined by **Lue et al., (2008)**.

Calculation of Pollution Indices

In an attempt to understand the pattern of metal contamination in the area, useful tools including pollution indices such as; i)

Geo-accumulation Index (Igeo), ii) Enrichment Factor (EF), iii) Pollution Index (PI), and iv) Load Pollution Index (LPI) were used to calculate heavy metals concentration in soil.

Geo-accumulation Index

Geo-accumulation index serves to assess contamination by comparing current and pre-industrial concentration of heavy metals (Muller, 1981). Background concentration of heavy metals in the earth's crust was used as a reference value implicating pre-industrial environment. Igeo is calculated through the following equation:

$$Igeo = \log_2 [C_n / 1.5B_n]$$

Where C_n is concentration of the element (n) in the soil sample and B_n is geochemical background value. The background values provided by Liao et al., 2004, were used to calculate the index of geo-accumulation. The constant 1.5 helps to analyze natural fluctuation between the content of a given substance in environment and very small anthropogenic influences. Muller (1981) has distinguished six classes of geochemical index as given in table (1).

Table 1: Igeo classes with respect to soil quality (Muller, 1981)

Class	Value	Soil Quality
0	$Igeo < 0$	Practically uncontaminated
1	$0 < Igeo < 1$	Uncontaminated to moderately uncontaminated
2	$1 < Igeo < 2$	Moderately contaminated
3	$2 < Igeo < 3$	Moderately to heavily contaminated
4	$3 < Igeo < 4$	Heavily contaminated
5	$4 < Igeo < 5$	Heavily to extremely contaminated
6	$5 < Igeo < 6$	Extremely contaminated

Enrichment Factor

The enrichment factor is established on the bases of standardization of a tested element against a reference one. A reference element is the one characterized by low occurrence variability such as Al, Fe, or Zn. In this study Al was used as a reference element since its concentration in earth's crust is low. The enrichment factor was calculated using the formula based on the equation suggested by Buat-Menard (1979):

Where C_n is the content of the examined element in the investigated area, C_{ref} is the content of the reference element in the same area, B_n is the content of the examined element in the reference location and B_{ref} is the content of the reference element in reference location. Five contamination

categories are recognized on the basis of the enrichment factor (Faiz et al., 2009) as present in Table (2).

Pollution Index

The assessment of the soil contamination was also carried out using the pollution index PI and Pollution Load Index PLI. The contamination factor was calculated using the relation described by Hakanson (1980);

$$PI = C_n / C_b$$

Where C_n is the mean content of metals from at least five sampling sites and C_b is the pre-industrial concentration of individual metal. Four categories of pollution index were defined by (Hakanson, 1980) as given in Table (2).

Table 2: Categories of EF (Sutherland, 2000) and PI (Hakanson, 1980).

EF	Category	PI	Category
EF < 2	Deficiency to mineral enrichment	PI < 1	Low contamination factor indicating low contamination
EF < 2-5	Moderate enrichment	1 < PI < 3	Moderate contamination factor
EF < 5-20	Significant enrichment	3 < PI < 6	Considerable contamination factor
EF < 20-40	Very high enrichment	6 < PI	Very high contamination factor

Load Pollution Index

Each sampling site was evaluated for the extent of the metal pollution by employing the method based on the pollution load index (PLI) developed by Tomlinson et al. as follows:

$$PLI = (PI_1 * PI_2 * PI_3 *PI_n)^{1/n}$$

Where n is the number of metals studied (4 in this study) and PI is the pollution index as mentioned above. The PLI provides simple but comparative means for assessing a site quality, where a value of PLI < 1 denotes perfection; PLI = 1 denotes that only baseline levels of pollutants are present

and PLI > 1 would indicate deterioration of site quality.

RESULT AND DISCUSSION

Heavy metal concentrations in surface Soil and their characteristics

The range and mean concentration of Cd, Cu, Pb and Zn in surface soil samples downwind the phosphate fertilizer plant are summarized in Table 4, where concentration of heavy metal compared with sampling sites is illustrated in Fig. (2).

Table 4: Total metal concentration (mg/kg) of surface soil samples near the fertilizer plant

Element	Min		Average		Max		Stdv		Quartile_1		Quartile_2	
	Down*	Up#	Down	Up	Down	Up	Down	Up	Down	Up	Down	Up
Zn	68.82	11.10	86.00	11.31	131.34	11.58	13.16	0.25	77.19	11.18	93.24	11.42
Cu	6.33	9.13	20.75	9.27	35.13	9.45	7.73	0.16	14.03	9.18	26.72	9.34
Pb	0.01	5.11	3.57	5.17	9.84	5.22	2.07	0.06	2.40	5.14	4.24	5.20
Cd	0.01	0.31	0.06	0.34	0.83	0.36	0.18	0.03	0.00	0.33	0.00	0.36

*Up: samples taken from upwind area (reference samples)

*Down: samples taken from downwind area (Polluted area)

The mean value for Cd in surface soils is 0.056 mg/kg with a range from 0.0125 to 0.825 mg/kg which is in sub-levels of the world average of 10 mg/kg (US-EPA) as presented in Table (5).

Table 5: Metal concentration standards (mg/kg) compared to the reference samples

Standard	Cd	Cu	Pb	Zn
US/EPA	-	30	10	NR
GLC	-	NR	20	NR
WHO	0.3	4	20	50
FAO	0.01	0.20	5.00	2.00
Present study	0.34	9.27	5.17	11.31

US/EPA = United States Environmental Protection Agency.
WHO=World Health Organization

GLC = Great London Council.
FAO = Food and Agriculture Organization (United Nations).

Copper concentrations is 20.612 mg/kg and ranges from 12.665 to 35.132 mg/kg. However the concentration of Cu in control soils as shown from table 3 ranged from 9.13-9.45 mg/kg which is under the level of the world guide lines admitted by WHO. The mean value of Pb in soil samples is 3.57 mg/kg with a range of 0.01 to 9.84 mg/kg compared to the mean value for the control value (5.16 mg/kg), exceeds the Food and Agriculture Origination (FAO) limit is 5.00 mg/kg. The mean value of Zn in samples is 85.873 mg/kg with a range from 75.156 to 131.342 mg/kg was significantly higher than that in the control area (11.31 mg/kg), as presented in Table 6. However, some soil samples of Zn showed elevated levels of Zn with a maximum of 131.34 mg/kg. From soil sample analysis physicochemical properties were determined and presented in Table 7.

Table (6): Control values of heavy metals (mg/kg) of soil samples

Sample No.	Cd	Cu	Pb	Zn
1	0.35	9.45	5.22	11.25
2	0.31	9.23	5.17	11.58
3	0.36	9.13	5.11	11.10
Average	0.34	9.27	5.16	11.31

Table 7: The physicochemical properties of soil samples

Samples Parameter	pH (pH unit)	EC (µs/cm)	T.O.C (mg/kg)
Range	5.15 – 5.58	165 – 201	0.38 – 0.54
Control	7.02	85.50	0.34

For pH values there was a slight acidity which may contribute in heavy metal leach in soils causing ground water pollution. As for Total Organic Carbon (T.O.C) there was no significant variation in the values of (T.O.C) obtained from all analyzed soil samples. Total Organic Carbon values ranged between 0.38-0.54 mg/kg for all soil samples indicating that studied soil samples contain almost the same proportions in them. Results of conductivity measurements were quite high with values ranging from 165-201 Ms/cm. This may be due to the increase in the concentration of some soluble salts in the soils. Figure 3 represents concentrations of heavy metal against sampling sites.

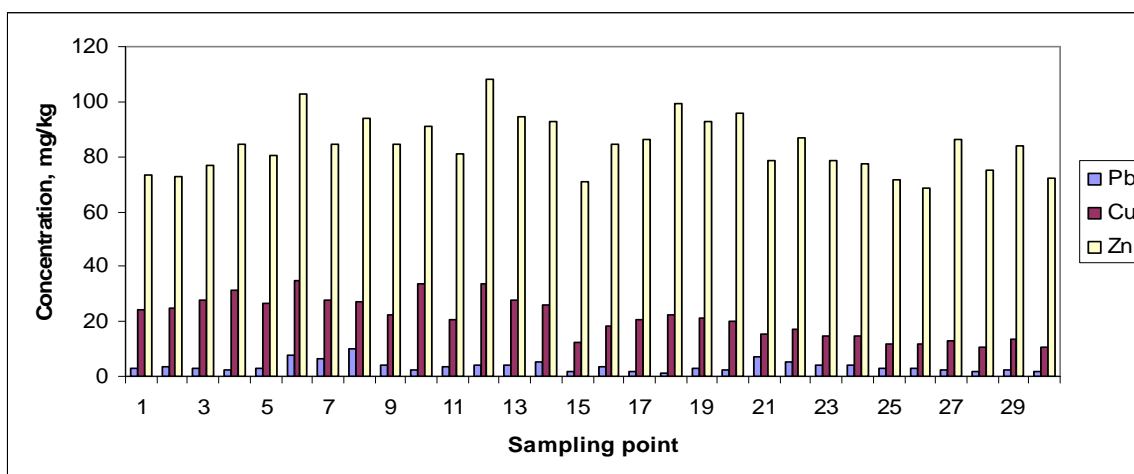


Fig.3: Concentration of heavy metals in different sampling sites.

Assessment of pollution Indices in Soil

The calculated results of I_{geo} of heavy metals in soil in the investigated area around the phosphate fertilizer factory are assessed and presented in Table 8. The I_{geo} ranges from (- 2.35) to (- 0.65) with a mean value of - 0.61 for Cd, 1.14 to 1.34 with a mean value of 0.48 for Cu, - 9.61 to 0.35 with a mean value of - 1.52 for Pb and 2.03 to 3.00 with a mean value of 2.33 for Zn.

Table 8: Assessment of Pollution index related to heavy metal in soils in the vicinity of the fertilizer plant.

	Enrichment factor EF		Pollution index PI		Igeo Geoaccumulation factor	
	Range	Mean	Range	Mean	Range	Mean
Cd	0.01- 0.05	0.03	0.29- 2.35	1.28	-2.35- 0.65	0.61
Cu	0.10- 0.57	0.34	1.02-37.05	3.41	1.14- 1.34	0.48
Pb	0.03- 0.30	0.11	0.012-1.91	0.69	-9.61- 0.35	-1.52
Zn	0.91-1.74	1.13	6.08-11.61	7.57	2.03- 3.00	2.33

The mean value of I_{geo} decreased in the order of Zn > Cu > Pb > Cd. The investigated area is slightly polluted with Cd and Pb, but Cu polluted the area moderately. The I_{geo} range of Zn pollution level lies in the fourth class indicating that the area is affected with high contamination of Zn element. The values of I_{geo} of the present study are compared with those reported in a number of previous studies as shown in table 9. In one of the previous studies in Gebze industrial area, in printing, leather and textile in Turkey, the highest I_{geo} values for metals were found as 10.2 Cd, 8.38 (Pb), 6.64 (Zn), and 3.63 (Cu) . Around cement factory in Ghana, the I_{geo} value of Cu was found to be 1.36, where I_{geo} values for fertilizer industry in India were 0.35 and 0.17 for Pb and Zn, but in fertilizer industry in Turkey, the Igeo value of Cadmium has reached 2.49 and 1.32 for Pb (Ahiamadjie *et al.*, 2011).

Table 9: Comparison of the geo-accumulation index calculated for the soil bound metals affected by industrial activities in different countries (17).

Order	Industry type	Country	Pb	Zn	Cu	Cd
1	Pb and Z smelter	China	3.98	2.67	0.19	4.72
2	Smelter	Belgium	4.91	--	3.57	4.74
3	Copper mines	Peru	2.38	0.72	--	8.67
4	Smelters, mining	Albania	2.17	0.81	0.71	4.32
5	Smelters and mining	Spain	1.50	--	--	--
6	Cement factory	Jordan	0.87	--	--	3.47
7	Printing, leather, textile	Turkey	8.38	6.64	3.63	10.2
8	Fertilizer and plastic products	Nigeria	2.25	0.80	--	2.68
9	Casting, chemicals	India	1.12	1.14	1.52	0.64
10	Present work	Egypt	-1.52	2.33	0.48	0.61
11	Around cement factories	Ghana	-	-	1.36	-
12	Fertilizer industry	India	0.35	0.17	-	-
13	Fertilizer industry	Turkey	-	-	-	2.49

Enrichment factor of heavy metal were calculated for each soil sample relative to the background values of abundance of chemical elements in the continental crust, choosing Al as the reference element. The EF of Cd, Cu, Pb and Zn is in the range of 0.01 to 0.05, 0.10 to 0.57, 0.03 to 0.40 and 0.91 to 1.74 with a mean value of 0.03, 0.34, 0.11 and 1.13 respectively in table 8. Spatial variation of heavy metals in the investigated area is given in Fig.4 to show distribution of Cd, Cu, Pb and Zn in each site for each element.

The mean EF for Cd, Pb, and Cu is less or close to unity but in case of Zn it reached 1.13. This can be interpreted that Cd may be originated from parent soils and the area is slightly polluted with Cu and Pb, but moderately polluted by Zn. Hence, EF is an effective tool to differentiate a natural origin from anthropogenic sources. The calculated means of EF, Igeo and PI of heavy metal (Cd, Cu, Pb and Zn) in soil samples are presented in Fig5. The mean value of EF and PI decreased in the order of Zn > Cu > Pb > Cd and Igeo for Pb showed a negative value indicating that lead pollution is a background state.

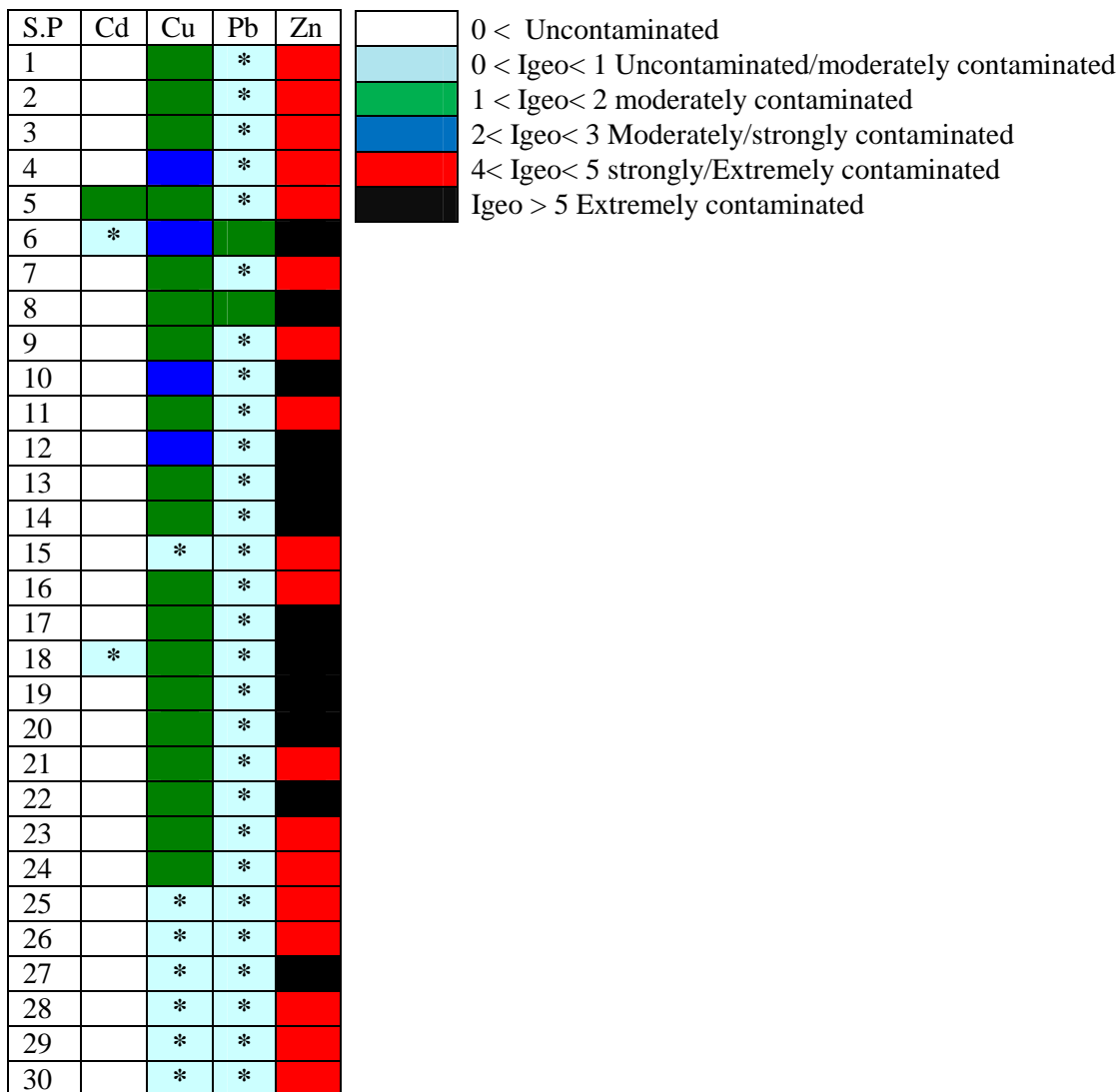


Fig.4: Spatial variation of heavy metal related to sampling sites

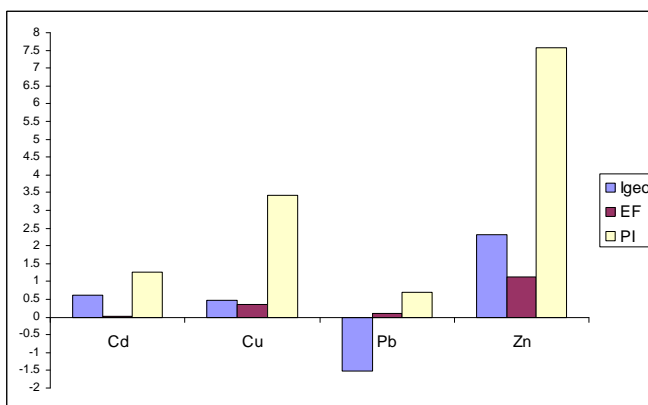
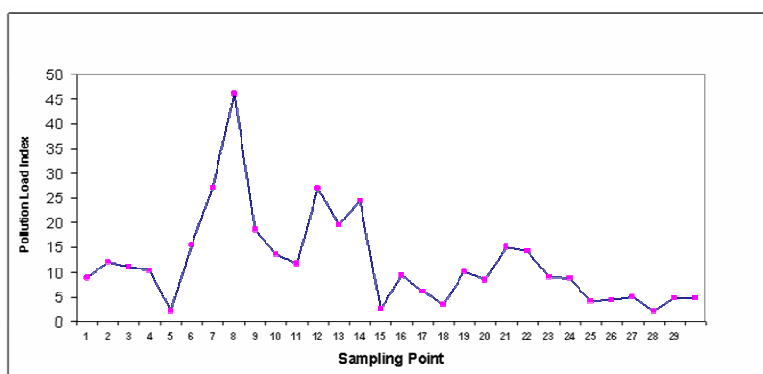


Fig. 5: The graph of mean values of EF, PI and Igeo against elements.

The PI is defined as the ratio of element concentration in soil samples to the background content of the abundance of chemical elements in the continental crust.



PLI < 0 Un polluted PLI = 1 Perfection PLI > 1 Deterioration of the environment

Fig. 6: PLI vs. sampling point

The pollution load Index PLI provides simple but comparative means for assessing a site quality, where a value of $PLI < 1$ denote perfection; $PLI = 1$ present that only baseline levels of pollutants are present and $PLI > 1$ would indicate deterioration of site quality (Tomlinson *et al.*, 1980) and coordination of PLI against sampling sites is expressed by Fig. 6.

In case of Cadmium the maximum range reached 2.35 but for Lead is 1.91 and the two values agree with the second class ($1 < PI \leq 3$), indicating a moderately pollution of the area. As for Copper the maximum value equals 37.05 and Zinc is 11.61. Hence, Copper and Zinc cause highly pollution to the investigated area. Calculation showed that (PLI) range is 0.21– 5.26 with a mean value of 2.17 indicating that the concerned heavy metals lie on the class $PLI > 1$ the high pollution degree of the investigated area.

CONCLUSIONS

Analysis of soil samples nearby phosphate fertilizer manufacturing plants showed that the adjacent area was highly enriched with heavy metals such as Cd, Cu, Pb and Zn. Obtained concentrations of these heavy metals found that they exceeded much more the concentrations of reference samples taken from unpolluted area (Thabet *et al.*, 2013) and also exceeded the limits of guidelines stated by World Health Organization WHO and Food and Agriculture Organization FAO.

The calculated results of geo-accumulation index for heavy metals in soil showed that Cd, Cu and Zn possess higher indices due to complex structures with soil constituents, but a negative index for Pb indicating that lead presents in its mobile fractions in soil solution. Enrichment factors showed that the area was moderately polluted with heavy metals but strongly polluted with contamination factors and this shows that there is a high contamination degree due to higher measured values of heavy metal concentrations than that of background concentrations.

Application of pollution indices gave an indicator that heavy metals present in the investigated soil with high levels representing a hazardous situation to the environment and the emissions of the phosphate fertilizer plants are the main source of contamination of the adjoining areas. This leads to the suggestion that phosphate fertilizer plant ought to apply control measures stated by governmental and organizational bodies and polluted soil should be remediated.

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الملخص العربي

تطبيق مؤشرات التلوث بالمعادن الثقيلة لتقييم التلوث في التربة القريبة من مصانع الأسمدة الفوسفاتية - أسيوط - مصر

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تم استخدام مؤشرات التلوث لقياس مستويات تركيز المعادن الثقيلة في المنطقة الواقعة تحت تأثير الانبعاثات الصادرة من مصانع الأسمدة الفوسفاتية بمنقباد بأسيوط ، حيث تنطلق من مختلف أوجه النشاط كميات ضخمة من الأتربة والأدخنة والغازات محتوية علي المعادن الثقيلة التي تترسب بفعل العوامل المناخية علي التربة المجاورة للمصانع و تسبب تلوثها بالمعادن الثقيلة . وقد تم تجميع عينات التربة من منطقة مجاورة و في اتجاه الريح من المصانع وتم تحديد إحداثيات مواقع العينات باستخدام النظام العالمي للإحداثيات (GPS) و تم تحليل العينات باستخدام جهاز مطياف الكتلة (ICP- MS) للوقوف علي تركيز هذه المعادن في منطقة الدراسة وبينت نتائج الدراسة ان المنطقة قد تأثرت كثيرا بالمعادن الثقيلة مثل الكاديوم والنحاس والرصاص والزنك ، حيث بلغ متوسط تركيزاتها (٠،٣ ، ٥٧،٠ ، ٩٤،٧ ، ٨٠،٧ مجم / كجم على الترتيب ، وهذه القيم زادت كثيرا عن المناسب الارضية (٠،٠١٣ مجم / كجم للكاديوم ، ٩،٦٢ مجم / كجم للنحاس ، ٥،١٧ مجم / كجم للرصاص وكذلك ١١،٥٦ مجم / كجم للزنك) والتي تم الحصول عليها من تحليل عينات للتربة تبعد مسافة ٥ كيلومترات عن منطقة التلوث و ضد اتجاه الريح ، وكذلك زادت هذه القيم المقاسة عن المعدلات العالمية لتلوث التربة والتي وضعتها منظمة الصحة العالمية (WHO) . كما تم تطبيق النمذجة الرياضية لمدلولات التلوث لتقييم تلوث التربة بالمعادن الثقيلة مثل: معامل التراكم الحيولوجي ومعامل التخصيب ومعامل الحمل البيئي ، وقد أظهرت حسابات درجة التلوث ان التربة في منطقة الدراسة تقع في التصنيف عالي التلوث من الكاديوم والنحاس والرصاص والزنك، وعلي ذلك فان الدراسة توصي الجهات المختصة بضرورة تطبيق اشتراطات التحكم في هذه الانبعاثات والزام هذه المصانع بالمستويات المسموح بها طبقا لقانون البيئة المصري رقم ٤ لسنة ١٩٩٤ وكذلك الاشتراطات الدولية في هذا الشأن ، وكذلك استخدام الطريقة المناسبة لمعالجة التربة الملوثة .