

## تقييم التلوث بالمعادن الثقيلة في التربة الرملية المروية بمياه الصرف الصحي

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### الملخص العربي

ان الهدف الرئيسي من هذه الدراسة تقييم تلوث التربة الرملية بالمعادن الثقيلة نتيجة استخدام مياه الصرف الصحي كمصدر للري ولمدد مختلفة. ولذلك تم اختيار مزرعة الجبل الاصفر وهي تمثل الارض الرملية . و تم اختيار اربع قطاعات تمثل فترات زمنية مختلفة لاستخدام مياه الصرف الصحي ( ١٠ سنوات)، ( ٢٠ سنة)، ( ٣٠ سنة) والارض البكر كمقارنة وتم جمع العينات علي اعماق (٠-٣٠ سم)،(٣٠-٦٠ سم. علاوه علي ذلك فأنه تم تقييم تأثير استخدام مياه الصرف الصحي علي تلوث التربة بتطبيق بعض المعادلات الرياضية وتشمل معامل التراكم الجيولوجي، معامل الثراء، معامل التلوث، درجة التلوث. أوضحت النتائج أن المحتوى الكلي من العناصر الثقيلة في التربة تباينت كمياتها نتيجة لاختلاف مدة الري (مستوي التلوث) الذي تعرضت له التربة .

كما اظهرت النتائج أن المحتوى الكلي لمعظم العناصر تحت الدراسة تقل بزيادة عمق طبقات الارض. وعند مقارنة النتائج المتحصل عليها مع الحدود المسموح بها لتراكم هذه العناصر في التربة وجد ان تركيز عنصر الكاديوم كان عند الحد المسموح به، بينما تركيز عنصر النحاس كان اكبر من الحد المسموح به، في حين ان تركيزات عناصر المنجنيز و النيكل والرصاص والزنك لم تتجاوز الحدود المسموح بها. أوضحت المعادلات الرياضية المستخدمة لتقييم درجة التلوث بالمعادن الثقيلة أن معامل الثراء (EF) لجميع العناصر تحت الدراسة سجلت قيما أقل من ٢ وبالتالي تقييم التربة علي ان هناك نقص او انخفاض في معدل الثراء. كما سجلت قيم معامل التراكم الجيولوجي (Igeo) قيما اقل من صفر لجميع العناصر تحت الدراسة وبالتالي يمكن تقييم التربة علي انها غير ملوثة . في حين أظهرت النتائج أن معامل التلوث (C<sub>f</sub>) لجميع العناصر ماعد الرصاص تحت الدراسة كانت تقل مع زيادة عمق القطاع الارضي ، ومن ناحية أخرى كانت قيم عناصر (النحاس، المنجنيز، النيكل، الرصاص، الزنك) أقل من (1 < C<sub>f</sub>) وبالتالي تقييم التربة علي انها غير ملوثة بهذه العناصر، بينما كانت قيم عناصر ( الكاديوم و الحديد) تقع ما بين (3 < C<sub>f</sub> < 1) وبالتالي تقييم التربة علي انها معتدلة التلوث بهذه العناصر. بالنسبة لدرجة تلوث التربة (C<sub>d</sub>) أوضحت النتائج أن جميع قيم العناصر تحت الدراسة تقل مع زيادة استخدام مياه الصرف الصحي في الري وكذلك بزيادة عمق القطاع الارضي ، كما كانت جميع القيم (C<sub>d</sub> < 1.5) وبالتالي تقييم التربة علي انها عديمة او قليلة في درجة التلوث لجميع العناصر تحت الدراسة.

## HEAVY METALS POLLUTION ASSESMENT IN SANDY SOILS IRRIGATED WITH SEWAGE WATER

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**ABSTRACT:** Seven heavy metals (Cd, Cu, Fe, Cu, Mn, Ni, Pb and Zn) were analyzed in Soil samples. The results showed that the total content of heavy elements in the soil ranged from 2.02 to 2.74 mg / kg for cadmium, from 159.785 to 248.77 mg / kg of copper, from 4546 to 7268.55 mg / kg of iron, from 65.43 to 226.89 mg / kg of manganese, from 41.89 to 57.06 mg / kg of nickel, from 55.51 to 72.46 mg / kg of lead, from 105.428 to 190.82 mg / kg of zinc. Results indicated that the concentration of the Cd is in the critical, the concentration of Cu is higher than critical level and Mn, Ni, Pb and Zn concentrations were in the permissible levels. Enrichment Factors (EF) of the heavy metals in soil calculated ranged from 0.362 to 1.52. The Geo-accumulation Index ( $I_{geo}$ ) calculated gave values indicating no pollution in sandy soil and ranged from -1.052 to -0.619. While the Contamination factor ( $C_f$ ) ranged from 0.84 to 1.26, on the other hand Cd, Fe and Pb were described as moderate contamination while other metals described as low contamination. Degree of contamination ( $C_d$ ) ranged from 0.85 to 1.35 with an average 0.95 these values were nil to very low degree of contamination.

**Key words:** Heavy metal, Soil, Enrichment factor (EF), Geoaccumulation Index ( $I_{geo}$ ), Contamination factor ( $C_f$ ) and Degree of contamination ( $C_d$ ).

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### INTRODUCTION

Pollution of the natural environment by heavy metals is a worldwide problem because these metals are indestructible and most of them have toxic effects on living organisms, when they exceed a certain concentration (Chen, *et al.*, 2007). Heavy metals are of high ecological significance since they are not removed from water as a result of self purification, but accumulate in reservoirs and enter the food chain (Loska and Wiechula, 2003). There is increasing awareness that heavy metals present in soil may have negative consequences on human health and on the environment (Abrahams, 2002; Schroeder *et al.*, 2004 and Selinus *et al.*, 2005). From the environmental point of view, all heavy metals are important because they can not be biodegraded and are largely immobile in the soil system, so they tend to accumulate and persist in urban soils for a long time. This results in levels that are harmful to humans upon both acute and chronic exposure (Brinkmann, 1994; Sheppard, 1998). The most frequently reported heavy

metals with regards to potential hazards and the occurrence in contaminated soils are Cd, Ni, Mn, Pb, Zn, Fe and Cu (Alloway, 1995). The concentration of these toxic elements in soils may be derived from various sources, including anthropogenic pollution, weathering of natural high background rocks and metal deposits (Senesi *et al.*, 1999). Contamination of soils by heavy metals is the most serious environmental problem and has significant implications for human health. Soils are generally regarded as the ultimate sink for heavy metals discharged into the environment and many heavy metals are bound to soils. Part of the difficulty with heavy metals studies lies in the complex nature of soils (Dang *et al.*, 2002; Obiajunwa *et al.*, 2002). Anthropogenic inputs of heavy metals are associated with industrialization and agricultural practices. Sources such as atmospheric deposition, waste disposal, waste incineration, urban effluent, traffic emissions, fertilizer application and long-term application of wastewater in agricultural land constitute the major anthropogenic

inputs. Metals from anthropogenic sources tend to be more mobile than those from pedogenic or lithogenic sources (Chlopecka *et al.*, 1996). Generally the distribution of heavy metals is and influenced by the nature of parent materials, climatic conditions and their relative mobility depending on soil parameters, such as mineralogy, texture and classification of soil (Krishna and Govil, 2007). Some physicochemical properties of soils such as pH and Organic carbon (OC) are important parameters that control the accumulation and the availability of heavy metals in the soil environment (Einax and Soldt, 1999). Therefore, the objective of this study was to determine the concentration of heavy metals and assessment of metals concentration of sandy soil as a result of sewage water application for long periods.

**MATERIALS AND METHODS**

**Collection and preparation of samples:** Total 12 soil samples were collected at El-Gabal El-Asfar farm at north greater Cairo, Egypt. This area is well characterized by using sewage water as a

source of irrigation. Four soil profiles were chosen from each location to representative the three periods of sewage water utilization. Profile1 (P<sub>0</sub>): Non polluted soils from El-Gabal El-Asfar farm (Non Cultivated). Soil samples for this profile taken at depths of 0-30, 30-60 cm, where the deeper layer was solid rock. Profile 2 (P<sub>10</sub>): Polluted soils from El-Gabal El-Asfar farm (Cultivated and subjected to swage effluent irrigation more than 10 years). Profile 3 (P<sub>20</sub>): Polluted soils from El-Gabal El-Asfar farm (Cultivated and subjected to swage effluent irrigation more than 20 years). Profile 4 (P<sub>30</sub>): Polluted soils from El-Gabal El-Asfar farm (Cultivated and subjected to swage effluent irrigation more than 30 years). Soil samples for this profile ((P<sub>10</sub>, P<sub>20</sub> and P<sub>30</sub>) taken at depths of 0-30, 30-60 and 60-90 cm, where the deeper layer was ground water. The samples were arid-dried, crushed, passed through a 2mm sieves, mixed thoroughly and stored in clean suitable plastic bags until they were analyzed Table (1)

shows some physical and chemical properties of the tested soil sample.

**Table (1) some physical and chemical properties of studied soil profile.**

| Pollution time (year) | Depth (cm) | Particle size distribution % |      |      | Soil texture | pH 1:2.5 | EC $\mu\text{mhos cm}^{-1}$ | CaCO <sub>3</sub> % | OM %  | CEC meq/100g soil |
|-----------------------|------------|------------------------------|------|------|--------------|----------|-----------------------------|---------------------|-------|-------------------|
|                       |            | Sand                         | Silt | Clay |              |          |                             |                     |       |                   |
| 0 (P <sub>0</sub> )   | 0-30       | 97.75                        | 2.25 | 0.00 | Sandy        | 7.49     | 775.0                       | 2.80                | 0.31  | 2.25              |
|                       | 30-60      | 97.0                         | 3.0  | 0.00 | sandy        | 7.64     | 685.0                       | 2.80                | 0.30  | 2.30              |
| Mean                  |            | 97.38                        | 2.63 | 0.00 |              | 7.57     | 730.0                       | 2.80                | 0.305 | 2.28              |
| 10 (P <sub>10</sub> ) | 0-30       | 96.0                         | 2.0  | 2.00 | Sandy        | 7.10     | 139.1                       | 2.91                | 2.15  | 3.0               |
|                       | 30-60      | 97.45                        | 2.55 | 0.00 | Sandy        | 7.50     | 116.5                       | 2.80                | 1.91  | 2.10              |
|                       | 60-90      | 97.7                         | 2.3  | 0.00 | Sandy        | 7.60     | 264.0                       | 2.80                | 1.91  | 1.86              |
| Mean                  |            | 97.05                        | 2.28 | 0.67 |              | 7.4      | 173.2                       | 2.84                | 1.99  | 2.32              |
| 20 (P <sub>20</sub> ) | 0-30       | 85.5                         | 2.5  | 2.00 | Sandy        | 6.68     | 141.8                       | 2.8                 | 2.27  | 2.42              |
|                       | 30-60      | 97.5                         | 2.5  | 0.00 | Sandy        | 7.0      | 127.2                       | 2.8                 | 2.02  | 1.66              |
|                       | 60-90      | 97.5                         | 2.5  | 0.00 | Sandy        | 7.20     | 84.4                        | 2.74                | 1.90  | 1.76              |
| Mean                  |            | 93.5                         | 2.5  | 0.67 |              | 6.96     | 117.8                       | 2.78                | 2.06  | 1.95              |
| 30 (P <sub>30</sub> ) | 0-30       | 95.2                         | 2.3  | 2.50 | Sandy        | 6.40     | 138.9                       | 2.74                | 2.38  | 3.52              |
|                       | 30-60      | 96.4                         | 2.6  | 0.00 | Sandy        | 6.50     | 90.3                        | 2.80                | 2.15  | 3.48              |
|                       | 60-90      | 97.5                         | 2.5  | 0.00 | Sandy        | 6.91     | 97.3                        | 2.69                | 2.03  | 3.97              |
| Mean                  |            | 96.36                        | 2.46 | 0.83 |              | 6.60     | 108.83                      | 2.74                | 2.18  | 3.65              |

**Method used for analysis:**

Approximately weight 0.25g of the soil sample was again treated with 9 ml concentrated nitric acid (HNO<sub>3</sub>) and 3 ml concentrated HCl, put in MF/HV vessels of microwave digester according to (ISO, 2002 and EPA, 1992). Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) model 5300 DV Optima Perkin Elmer, was used for determination of the total concentration of the seven heavy metals under investigation (Cd, Cu, Fe, Mn, Ni, Pb and Zn).

**Quality Assurance:** An appropriate quality assurance procedure was carried out to ensure reliability of the results. Samples were generally carefully handled to avoid contamination. All glassware before used were washed with distilled water, soaked in nitric acid (30%) overnight, rinsed in deionized water and air-dried. Double distilled deionized water was used throughout the study. Chemicals, standard solutions and other reagents were obtained from Fluka/Merk and were of analytical grade. Reagent blank determinations were used to correct the instrument reading. Average values of three replicates were taken for each element determination. All readings were background corrected; calibration was done before measurement of samples using series of standard solutions containing mixtures of the various elements. Quality control samples were checked every 10 samples. For validation of the analytical procedure the average recovery rates of studied metals were within 90±10% (Riffat *et al.*, 2010 and Mohamed *et al.*, 2006).

**Statistical Analysis:** Statistical Analysis was done for all variables. Regression analysis was performed to assess the association between metal fraction, soil properties and irrigation time. All statistical analysis were carried out by using procedures available in the SPSS v.10 (SPSS Inc., Chicago, IL, USA) Statistical

Package for Social Science computer program.

**RESULTS AND DISCUSSION**

The total metal concentrations and average values for each sampling site found in soils are shown in Table (2). Metal contents were ranging over following intervals: Metal contents were ranging over following intervals: Cd: 2.025-2.738mg.kg<sup>-1</sup>; Cu: 159.785-248.77mg.kg<sup>-1</sup>; Fe: 4546.5–7268.55mg.kg<sup>-1</sup>; Mn: 65.43-226.88 mg.kg<sup>-1</sup>; Ni: 41.89-57.06 mg.kg<sup>-1</sup>; Pb: 55.51-72.45 Zn: 105.428-190.81 mg.kg<sup>-1</sup>. Mean contents of the ecosystem studied were: Cd: 2.025 mg.kg<sup>-1</sup>; Cu: 159.785 mg.kg<sup>-1</sup>; Fe: 5552.15 mg.kg<sup>-1</sup>; Mn: 131.47 mg.kg<sup>-1</sup>; Ni: 47.72 mg.kg<sup>-1</sup>; Pb: 61.22 mg.kg<sup>-1</sup>; Zn: 156.09 mg.kg<sup>-1</sup>. Meanwhile, the comparison of the experimentally total concentration means of the selected heavy metals in the cultivated soils with those in uncultivated (P<sub>0</sub>) showed that the higher concentrations of Cd, Mn, and Pb were increased after 10 years (P<sub>10</sub>) and then decreased by increasing irrigation periods (P<sub>20</sub> and P<sub>30</sub>). On the other hand, the higher determined total concentration values of Cu and Zn were reached in the studied soils after 20 years (P<sub>20</sub>), but Fe accumulated after 30 years (P<sub>30</sub>). It was observed that the concentrations of Ni were decreased by increasing the periods of sewage water application. The comparison of the average total concentration of heavy metals in soil Table (2) with the permissible level (Alloway, 1990; Kabata-Pendias and Pendias, 1992) shows that the concentration of Cd is in the critical , the concentration of Cu is higher than critical level and Mn, Ni, Pb and Zn concentration was in the permissible levels. These results can be interpreted on the basis of the difference in soil chemical properties and their relationships with the examined metals. The equations which describe the relationships between the irrigation periods (years) and the total experimentally concentration values are shown in Table (3).

**Table (2): Effect of sewage irrigation periods (years) and soil profile depth (cm) on the total concentration of studied heavy metals in El Gabal El Asfar soils.**

| POLLUTION TIME (YEAR) | PROFILE DEPTH (CM) | TOTAL HEAVY METALS (MG.KG <sup>-1</sup> ) |         |          |           |        |         |         |
|-----------------------|--------------------|---|---------|----------|-----------|--------|---------|---------|
|                       |                    | CdT                                       | CuT     | FeT      | MnT       | NiT    | PbT     | ZnT     |
| 0 (P <sub>0</sub> )   | 0-30               | 2.304                                     | 245.437 | 4935.860 | 157.178   | 44.541 | 67.556  | 196.937 |
|                       | 30-60              | 2.353                                     | 242.792 | 4157.210 | 125.130   | 69.580 | 54.909  | 164.926 |
| Mean                  |                    | 2.329                                     | 244.115 | 4546.535 | 141.154   | 57.061 | 61.233  | 180.932 |
| 10 (P <sub>10</sub> ) | 0-30               | 3.506                                     | 171.352 | 6230.470 | 243.592   | 43.884 | 76.884  | 193.779 |
|                       | 30-60              | 2.290                                     | 175.123 | 4069.980 | 357.960   | 43.746 | 80.986  | 179.802 |
|                       | 60-90              | 2.419                                     | 333.380 | 6099.810 | 79.109    | 38.045 | 59.500  | 130.984 |
| Mean                  |                    | 2.738                                     | 226.818 | 5466.753 | 226.887   | 41.892 | 72.457  | 147.188 |
| 20 (P <sub>20</sub> ) | 0-30               | 3.391                                     | 413.869 | 6064.700 | 91.403    | 50.662 | 83.064  | 252.862 |
|                       | 30-60              | 1.976                                     | 228.962 | 4414.040 | 46.871    | 46.345 | 44.583  | 167.570 |
|                       | 60-90              | 1.794                                     | 103.750 | 4301.530 | 58.020    | 36.319 | 38.896  | 152.015 |
| Mean                  |                    | 2.387                                     | 248.770 | 4926.757 | 65.431    | 44.442 | 55.514  | 190.816 |
| 30 (P <sub>30</sub> ) | 0-30               | 2.537                                     | 256.982 | 9943.400 | 109.899   | 58.201 | 52.258  | 111.338 |
|                       | 30-60              | 1.900                                     | 120.148 | 5978.510 | 52.889    | 41.818 | 35.119  | 79.737  |
|                       | 60-90              | 1.639                                     | 102.226 | 5883.740 | 114.525   | 42.536 | 79.696  | 125.208 |
| Mean                  |                    | 2.025                                     | 159.785 | 7268.55  | 92.348    | 47.518 | 55.691  | 105.428 |
| Average Mean          |                    | 2.370                                     | 219.872 | 5552.149 | 131.477   | 47.728 | 61.224  | 156.091 |
| Normal level          |                    | 0.01-2                                    | 2-250   | -        | 20-1000   | 2-750  | 2-300   | 1-900   |
| Critical level        |                    | 3-8                                       | 60-125  | -        | 1500-3000 | 100    | 100-400 | 70-400  |

**Table (3): Relationships between the irrigation periods (years) and the total experimentally concentration values.**

| METALS | EQUATIONS                                | R2    |
|--------|--|-------|
| Cd     | $Cd_T = -0.0023 p^2 + 0.0511 p + 2.3595$ | 0.947 |
| Cu     | $Cu_T = -0.2312 p^2 + 4.0018 p + 235.57$ | 0.815 |
| Fe     | $Fe_T = 2.4163 p^2 - 9.8798 p + 4740.9$  | 0.744 |
| Mn     | $Mn_T = -0.1459 p^2 + 1.3126 p + 162.95$ | 0.369 |
| Ni     | $Ni_T = 0.0423 p^2 - 1.5702 p + 56.135$  | 0.872 |
| Pb     | $Pb_T = -0.018 p^2 + 0.3203 p + 63.689$  | 0.236 |
| Zn     | $Zn_T = -0.1884 p^2 + 3.6515 p + 174.83$ | 0.821 |

T = total concentration (mg.Kg<sup>-1</sup>)      p = periods of irrigation (years)

According to the correlation coefficients, results pointed out that quadratic equations model are the better to explain the relationships between total concentration values and the irrigation period with sewage. By applying quadratic equations model the

correlation coefficients were 0.947, 0.815, 0.744, 0.872 and 0.821 for Cd, Cu, Fe, Ni, and Zn, respectively but in the case of Mn and Pb the coefficients were 0.369 and 0.236 in the same order. The highest values of correlation coefficients in the case of Cd,

Cu, Fe, Ni, and Zn exhibited the superior relation between total concentration of these metals and the irrigation periods with sewage water. As regard to the effect of soil profile depth on the studied heavy metals obtained results placed in Table (2) illustrated that the total concentration values ( $\text{mg.Kg}^{-1}$ ) of Cd, Cu, Fe, and Mn were decreased generally by increasing the soil profile depth to the lowest values in the (90–120 cm). For Ni, Pb, and Zn metals the total concentration values ( $\text{mg.Kg}^{-1}$ ) were gradually decreased with increasing the soil profile depth to the lowest values at (60–90 cm) layer. These result comes in harmony with several others reported that the total content of some metals increased after irrigation with increasing the sewage effluent. However, the values obtained for most of the elements decreased with depth of soil profile. Saber,1986; Ashmawy,1988; El-Hassanin *et al.*, 1993; Abo El-Abbas,2001, Abd El-Shafy *et al.*, 2003 ; Kamel and Husien., 2007 and Soad *et al.*,2011; indicated that prolonging the irrigation periods increased the total and available form of some metals. Also concluded that the concentration of heavy elements have not accumulated to toxic levels even after 67 yrs with sewage irrigation. Total concentration can be arranged from higher to lower mean content in this area as:

$\text{Fe} > \text{Cu} > \text{Zn} > \text{Mn} > \text{Pb} > \text{Ni} > \text{Cd}$ .

### **Assessment of metals concentration:**

#### **1- Assessment According to Enrichment Factor (EF):**

A common approach to estimate how much the soil/sediment is impacted (naturally and anthropogenically) with heavy metal is to calculate the Enrichment Factor (EF) for metal concentrations above uncontaminated background levels (Huu *et al.*, 2010). Pollution will be measured as the amount or ratio of the sample metal enrichment above the concentration present in the reference station or material (Abraham and Parker, 2008 and Mediola *et al.*, 2008). The EF method normalizes the measured heavy metal content with respect to a samples reference such as Fe, Al, Zn, Sc, Ti and Si (Shotyk *et al* (2000), Hernandez *et al*

(2003) , Abraham and Parker (2008), Ata *et al* (2009) and Aikpokpodion *et al* (2010). The EF of a heavy metal in sediment can be calculated with the following Formula: (Huu *et al.*, 2010)

$$EF = \frac{\left(\frac{M}{Fe}\right)_{Sample}}{\left(\frac{M}{Fe}\right)_{Background}}$$

#### **Where:**

EF is the Enrichment Factor.

$[M/Fe]_{Sample}$  is the ratio of metal and Fe concentration of the sample.

$[M/Fe]_{Background}$  is the ratio of metal and Fe concentration of background.

Enrichment factor (EF) can be used to differentiate between the metals originating from anthropogenic activities and those from natural procedure, and to assess the degree of anthropogenic influence. Five contamination categories are recognized based on the enrichment factor as follows: (Sutherland, 2000) and Aikpokpodion *et al* (2010) Based on the Enrichment Factor (EF), the heavy metal contamination can be classified into the following levels: 1)  $EF \leq 2$ : Deficiency to minimal enrichment; 2)  $2 < EF \leq 5$ : moderate enrichment; 3)  $5 < EF \leq 20$ : significant enrichment; 4)  $20 < EF \leq 40$ : very high enrichment; 5)  $EF \leq 40$ : extremely high enrichment. In this study iron was used as reference values of heavy metals background concentrations to differentiate natural from anthropogenic components according to Rubio *et al.*, (2000) and Akoto *et al.*, (2008). The averages of Cd enrichment factor (EF) ranged from 0.55 to 0.991, from 0.40 to 0.903, from 0.42 to 1.523, from 0.55 to 0.742, from 0.77 to 0.87 and from 0.38 to 0.963 for Cd, Cu, Mn, Ni, Pb, and Zn, respectively. The higher (EF) values of all selective heavy metals were obtained after 10 years ( $P_{10}$ ) for Cd, Mn, and Pb but for Cu, Ni, and Zn were obtained after 20 years ( $P_{20}$ ). Meanwhile the lowest (EF) values were obtained after 30 years ( $P_{30}$ ) for all studied heavy metals. These results revealed that the (EF) values of the investigated heavy metals increased after 10 or 20 years irrigation with sewage water and

then decreased. The mean values of (EF) are arranged in the following order:  $P_{10} > P_{30} > P_{20}$  for Mn;  $P_{10} > P_{20} > P_{30}$  for Cd, and Pb; and  $P_{20} > P_{10} > P_{30}$  for Cu, Ni, and Zn. Generally, the calculated values of (EF) for the studied heavy metals were less than 2 based on the categorization of Aikpokpodion *et al* (2010). All soil heavy metals under study can be classified as deficiency to minimal enrichment. On the other hand, the enrichment factor values for each cultivated period can be arranged as the following order:

Cultivated soil for 10 years ( $P_{10}$ ): Mn > Cd > Pb > Zn > Cu > Ni. Cultivated soil for 20 years ( $P_{20}$ ): Zn > Cd > Cu > Pb > Ni > Mn. Cultivated soil for 30 years ( $P_{30}$ ): Pb > Ni > Cd > Mn > Zn > Cu.

## 2- Assessment according to Geoaccumulation Index ( $I_{geo}$ ):

A common criterion to evaluate the heavy metal pollution in soil is the geoaccumulation index ( $I_{geo}$ ) Hoda Ahdy *et al.*, (2009). This was originally defined by Muller (1979) to determine metals contamination in soils, by comparing current concentrations with background levels and can be calculated by

the following equation: 
$$I_{geo} = \log_2 \left[ \frac{C_n}{1.5 \times B_n} \right]$$

**Where:**  $C_n$  is the measured concentration of the examined metal (n) in the soil; and  $B_n$  is the geochemical background concentration value of the metals (n) in the background or control within the study area; Factor 1.5 is the background matrix correction factor due to lithogenic effects. (Aikpokpodion *et al.*, 2010). Muller (1981) based on the  $I_{geo}$  value, the degree of heavy metal contamination is classified as follows:

1)  $I_{geo} < 0$ : unpolluted; 2)  $0 \leq I_{geo} < 1$ : unpolluted to moderately polluted; 3)  $1 \leq I_{geo} < 2$ : moderately polluted; 4)  $2 \leq I_{geo} < 3$ : moderately to strongly polluted; 5)  $3 \leq I_{geo} < 4$ : strongly polluted; 6)  $4 \leq I_{geo} < 5$ : strongly to very strongly polluted; 7)  $I_{geo} \geq 5$ : very strongly polluted. According to the Muller Scale, the calculated results of geoaccumulation index ( $I_{geo}$ ) are illustrated in Fig. (2). The geoaccumulation index ( $I_{geo}$ )

were ranged from -0.810 to -0.38, -1.32, - to -0.764, -0.487 to -0.046, -1.752 to -0.154, -1.034 to -0.860, -0.809 to -0.355, and from -1.39 to -0.545 for Cd, Cu, Fe, Mn, Ni, Pb, and Zn respectively. On the basis of the mean values the effect of irrigation periods on ( $I_{geo}$ ) the soils are enriched in the following order:  $P_{10} > P_{20} > P_{30}$ . The largest values of the studied metals ( $I_{geo}$ ) were after 10 years and the lowest were obtained after 30 years for all investigated metals. Generally, the studied heavy metals had ( $I_{geo}$ ) less than  $\leq 0$  based on the categorization of Muller (1981). Accordingly, all heavy metals in the studied soils can be classified as class zero which considered as unpolluted soils. The effect of irrigation periods on the mean values of ( $I_{geo}$ ) the studied metals can be arranged as the following orders:

Cultivated soil ( $P_{10}$ ): Mn > Fe > Pb > Cd > Zn > Cu > Ni. Cultivated soil ( $P_{20}$ ): Fe > Zn > Cd > Cu > Pb > Ni > Mn. Cultivated soil ( $P_{30}$ ): Fe > Pb > Ni > Cd > Mn > Zn > Cu. The obvious orders revealed that Mn had the largest value of ( $I_{geo}$ ) after 10 years ( $P_{10}$ ) of irrigation with sewage water but Fe metal had the largest value of ( $I_{geo}$ ) after 20 and 30 years ( $P_{20}$  and  $P_{30}$ ). When discussing the impact of the depth of the soil profile on the values of the ( $I_{geo}$ ), the results in Fig. (3) shown that in general the values of ( $I_{geo}$ ) were decreased with the increase in the depth of soil profile.

## 3- Assessment According to Contamination Factor ( $C_f$ ) and Modified Degree of Contamination ( $C_d$ ):

The contamination factor ( $C_f$ ) and the degree of contamination ( $C_d$ ) are used to determine the contamination status of soil in the present study. Contamination factor ( $C_f$ ) values are suggesting for describe the contamination conditions of soils (Hakanson, 1980, Liu *et al.*, 2005, Ata, 2009 and Aikpokpodion, 2010). The metal contamination factor ( $C_f$ ) is defined as the following equation:

$$C_f = \frac{C_{0-1}}{C_n}$$

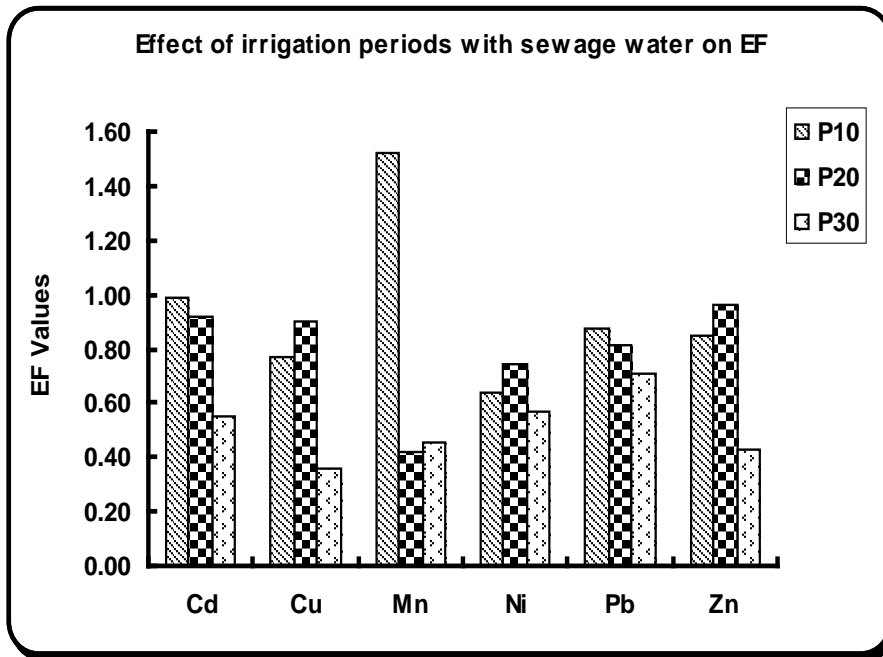


Fig. (1): Effect of sewage water irrigation periods (years) on enrichment factor (EF) of selected heavy metals of El Gabal El Asfar soils

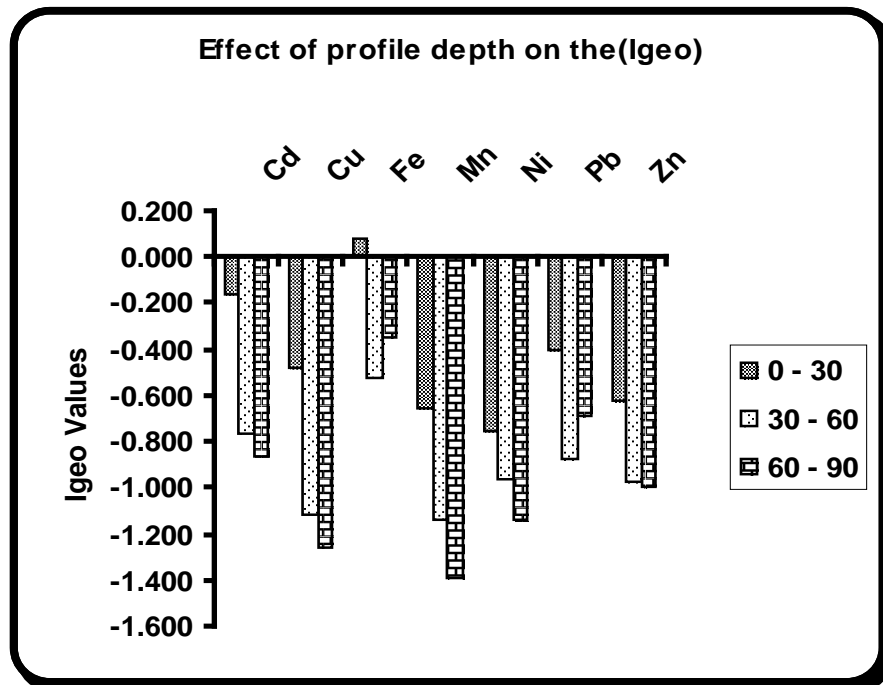


Fig. (2): Effect of sewage water irrigation periods (years) on geoaccumulation index(Igeo) of the selected heavy metals.



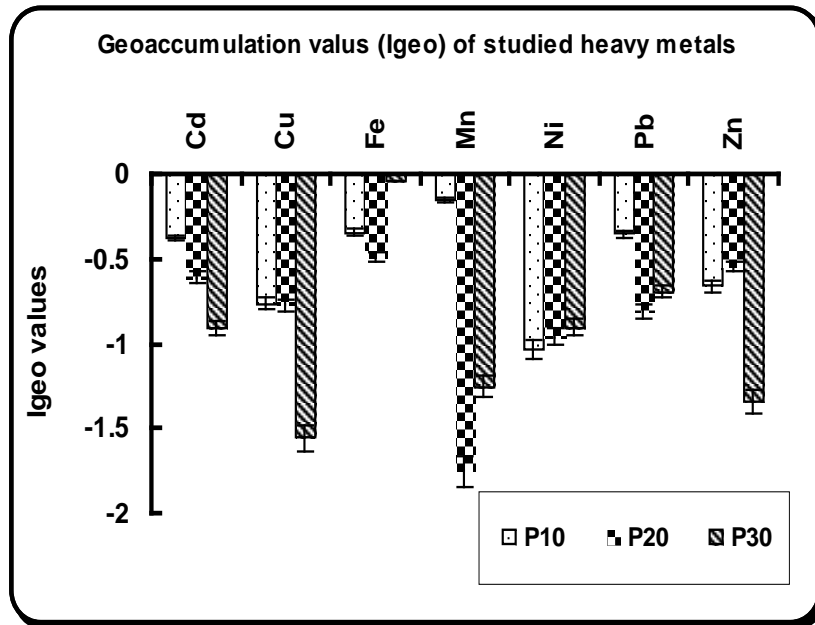


Fig. (3): Effect of soil profile depth on geoaccumulation index ( $I_{geo}$ ) of the selected heavy metals.

**Where:**  $C_f$  is the contamination factor; and ( $C_{0.1}$ ) is the metal concentration of the sample; the ( $C_n$ ) is the background value of the metal. The contamination factor ( $C_f$ ) is defined according to four categories (Liu *et al.*, 2005) as follows  $C_f < 1$  low contamination factor;  $1 < C_f < 3$  moderate contamination factor;  $3 < C_f < 6$  considerable contamination factor  $C_f > 6$  very high contamination factor.

The degree of contamination ( $C_d$ ) was defined as the sum of all contamination factors according to (Hoda Ahdy *et al.*, 2009). For the classification and description of the degree of contamination ( $C_d$ ) in soil the following gradations are proposed by (Abraham and Parker, 2008):  $C_d < 1.5$  Nil to very low degree of contamination;  $1.5 < C_d < 2$  Low degree of contamination;  $2 < C_d < 4$  Moderate degree of contamination;  $4 < C_d < 8$  High degree of contamination;  $8 < C_d < 16$  Very high degree of contamination;  $16 < C_d < 32$  Extremely high degree of contamination; and  $C_d \geq 32$  Ultra high degree of contamination. Results of the contamination factor ( $C_f$ ) and the degree of

contamination ( $C_d$ ) of heavy metals in the studied soil samples are presented in Table (4). The contamination factors of the studied heavy metals can be arranged as the following orders: Cd ( $P_{10} > P_{20} > P_{30}$ ); Pb and Mn ( $P_{10} > P_{30} > P_{20}$ ); Zn and Cu ( $P_{20} > P_{10} > P_{30}$ ); Ni ( $P_{30} > P_{20} > P_{10}$ ); Fe ( $P_{30} > P_{10} > P_{20}$ ).

Gained results as shown in Table (4) illustrated that the ( $C_f$ ) average of all investigated metals except Pb were decreased with increasing soil profile depth. On the other hand we found that ( $C_f$ ) values for all studied heavy metal were described as low contamination ( $C_f < 1$ ) except for Cd and Fe which were described as moderate contamination ( $1 < C_f < 3$ ) (Liu *et al.*, 2005). We can arrange the studied metals according  $C_f$  values as the following order: Fe > Pb > Cd > Mn > Zn > Cu > Ni. As to degree of contamination ( $C_d$ ) results placed in Table (4) revealed that there were decreasing in ( $C_d$ ) values by increasing both irrigation periods and soil profile depth. The results placed in Table (4) showed that ( $C_d$ ) values were ranged between 0.87 and 1.35

with an average 0.95. These values of ( $C_d$ ) were nil to very low degree of contamination as described by (Abraham and Parker, 2008). Correlation between the total concentration of selected heavy metals and some physicochemical properties of the studied soils:

Data in Table (5) showed that sand contents did not significantly correlated with

different selected heavy metals with exception of Cd and Cu. The correlation coefficients of total cadmium (Cd) and total copper (Cu) concentration with sand content were negative significance at 0.05 level (-0.642\* and -0.647\* respectively).

**Table (4): Effect of irrigation periods with sewage water and soil profile depth on contamination factor ( $C_f$ ) and degree of contamination ( $C_d$ ) of the selected heavy metals of El Gabal El Asfar soils.**

| IRRIGATION PERIODS (YEARS) | PROFILE DEPTH (CM) | CONTAMINATION FACTOR ( $C_f$ ) OF THE STUDIED HEAVY METALS |      |      |      |      |      |      | SUM OF $C_f$ | CON. DEGREE ( $C_d$ ) |
|----------------------------|--------------------|--|------|------|------|------|------|------|--------------|-----------------------|
|                            |                    | Cd   | Cu   | Fe   | Mn   | Ni   | Pb   | Zn   |              |                       |
| 10<br>( $P_{10}$ )         | 0-30               | 1.51   | 0.70 | 1.37 | 1.73 | 0.77 | 1.26 | 1.07 | 8.41         | 1.20                  |
|                            | 30-60              | 0.98   | 0.72 | 0.90 | 2.54 | 0.77 | 1.32 | 1.09 | 8.32         | 1.89                  |
|                            | 60-90              | 1.04   | 1.37 | 1.34 | 0.56 | 0.67 | 0.97 | 0.72 | 6.68         | 0.95                  |
| Means                      |                    | 1.18   | 0.93 | 1.20 | 1.61 | 0.73 | 1.18 | 0.96 | 7.80         | 1.35                  |
| 20<br>( $P_{20}$ )         | 0-30               | 1.46   | 1.70 | 1.33 | 0.65 | 0.89 | 1.36 | 1.40 | 8.77         | 1.25                  |
|                            | 30-60              | 0.85   | 0.94 | 0.97 | 0.33 | 0.81 | 0.73 | 0.93 | 5.56         | 0.79                  |
|                            | 60-90              | 0.77   | 0.43 | 0.95 | 0.41 | 0.64 | 0.64 | 0.84 | 4.66         | 0.67                  |
| Means                      |                    | 1.03   | 1.02 | 1.08 | 0.46 | 0.78 | 0.91 | 1.05 | 6.33         | 0.90                  |
| 30<br>( $P_{30}$ )         | 0-30               | 1.09   | 1.05 | 2.19 | 0.78 | 1.02 | 0.85 | 0.62 | 7.60         | 1.09                  |
|                            | 30-60              | 0.82   | 0.49 | 1.31 | 0.37 | 0.73 | 0.57 | 0.44 | 4.72         | 0.68                  |
|                            | 60-90              | 0.70   | 0.42 | 1.29 | 0.81 | 0.75 | 1.29 | 0.69 | 5.96         | 0.85                  |
| Means                      |                    | 0.87   | 0.65 | 1.60 | 0.66 | 0.83 | 0.91 | 0.58 | 6.99         | 0.87                  |
| Average Means              |                    | 1.02   | 0.87 | 1.29 | 0.91 | 0.78 | 1.02 | 0.87 | 7.60         | 0.95                  |

**Table (5): Correlation between physico chemical properties of El Gabal El Asfar soils and total concentration of the studied heavy metals.**

| ITEM              | CD <sub>T</sub> | CU <sub>T</sub> | FE <sub>T</sub> | MN <sub>T</sub> | NI <sub>T</sub> | PB <sub>T</sub> | ZN <sub>T</sub> |
|-------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Sand              | -0.642*         | -0.647*         | -0.262          | 0.106           | -0.237          | -0.339          | -0.561          |
| F (silt+clay)     | 0.743**         | 0.475           | 0.690*          | 0.082           | 0.490           | 0.196           | 0.292           |
| pH                | -0.038          | 0.076           | -0.666*         | 0.377           | -0.014          | 0.179           | 0.315           |
| EC                | 0.093           | 0.303           | -0.259          | 0.069           | 0.473           | 0.015           | 0.252           |
| CaCO <sub>3</sub> | 0.754**         | 0.374           | -0.116          | 0.391           | 0.086           | 0.061           | 0.476           |
| CEC               | -0.144          | -0.416          | 0.575           | -0.037          | 0.074           | 0.197           | -0.505          |
| O.M %             | 0.103           | -0.081          | 0.474           | -0.094          | -0.409          | 0.024           | -0.206          |

Concerning the correlation of fine particles (silt and clay) with total heavy metals concentration, there were no significance correlation except with Cd and Fe which were positive coefficient at 0.05 level 0.773\* and 0.690\* respectively. Results listed in Table (5) demonstrated that the correlation between soil pH and total heavy metals concentration did not significantly correlated except for Fe. The correlation coefficient of Fe with soil pH was high negative at 0.05 level and equal to -0.666\* (Rana *et al.*, 2010). In respect to relationship between soil calcium carbonate content there was no any correlations with the studied heavy metals except with Cd. The total Cd concentration was high bounded with soil CaCO<sub>3</sub> at 0.05 level and the coefficient correlation was positive significance (0.754\*\*) (Marija *et al.*, 2004).

### Conclusions

This survey has allowed us to determine total metals levels (Cu, Fe, Ni, Pb, Cd and Zn) in soils from the El-gabal El-Asfar farm on Egypt. Results indicated that the concentration of the Cd is in the critical, the concentration of Cu is higher than critical level and Mn, Ni, Pb and Zn concentrations were in the permissible levels. Soil pollution in the present study was assessed using enrichment factor, geoaccumulation index values, contamination factor and degree of contamination. The enrichment factor (EF) values showed that all soil heavy metals under study were less than 2 and can be classified as deficiency to minimal enrichment. The geoaccumulation index ( $I_{geo}$ ) allows us to conclude that, the concentrations of all heavy metals under study less than  $< 0$  and can be classified as class zero which considered as unpolluted soils. The contamination factors of the studied heavy metals found that ( $C_f$ ) values for all studied heavy metal were described as low contamination ( $C_f < 1$ ) except for Cd and Fe which were described as moderate contamination ( $1 < C_f < 3$ ). As to degree of contamination ( $C_d$ ) there were decreasing in ( $C_d$ ) values by increasing both irrigation periods and soil profile depth.

The values of ( $C_d$ ) were nil to very low degree of contamination. The correlation analysis of total heavy metals concentrations under study with soil properties showed that the increasing soil sand contents led to decrease both Cd and Cu. Total Fe and Cd concentration were increased with increasing fine soil particles. There was an inverse relationship between total concentration of iron and soil pH. The total cadmium concentration highly associated with soil CaCO<sub>3</sub> contents.

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## تقييم التلوث بالمعادن الثقيلة في التربة الرملية المروية بمياه الصرف الصحي

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### الملخص العربي

ان الهدف الرئيسي من هذه الدراسة تقييم تلوث التربة الرملية بالمعادن الثقيلة نتيجة استخدام مياه الصرف الصحي كمصدر للري ولمدد مختلفة. ولذلك تم اختيار مزرعة الجبل الاصفر وهي تمثل الارض الرملية . و تم اختيار اربع قطاعات تمثل فترات زمنية مختلفة لاستخدام مياه الصرف الصحي ( ١٠ سنوات)، ( ٢٠ سنة)، ( ٣٠ سنة) والارض البكر كمقارنة وتم جمع العينات علي اعماق (٠-٣٠ سم)،(٣٠-٦٠ سم. علاوه علي ذلك فأنة تم تقييم تأثير استخدام مياه الصرف الصحي علي تلوث التربة بتطبيق بعض المعادلات الرياضية وتشمل معامل التراكم الجيولوجي، معامل الثراء، معامل التلوث، درجة التلوث. أوضحت النتائج أن المحتوى الكلي من العناصر الثقيلة في التربة تباينت كمياتها نتيجة لاختلاف مدة الري (مستوي التلوث) الذي تعرضت له التربة .

كما اظهرت النتائج أن المحتوى الكلي لمعظم العناصر تحت الدراسة تقل بزيادة عمق طبقات الارض. وعند مقارنة النتائج المتحصل عليها مع الحدود المسموح بها لتراكم هذه العناصر في التربة وجد ان تركيز عنصر الكاديوم كان عند الحد المسموح به، بينما تركيز عنصر النحاس كان اكبر من الحد الحرج المسموح به، في حين ان تركيزات عناصر المنجنيز و النيكل والرصاص والزنك لم تتجاوز الحدود المسموح بها. أوضحت المعادلات الرياضية المستخدمة لتقييم درجة التلوث بالمعادن الثقيلة أن معامل الثراء (EF) لجميع العناصر تحت الدراسة سجلت قيما أقل من ٢ وبالتالي تقييم التربة علي ان هناك نقص او انخفاض في معدل الثراء. كما سجلت قيم معامل التراكم الجيولوجي (Igeo) قيما اقل من صفر لجميع العناصر تحت الدراسة وبالتالي يمكن تقييم التربة علي انها غير ملوثة . في حين أظهرت النتائج أن معامل التلوث (C<sub>f</sub>) لجميع العناصر ماعد الرصاص تحت الدراسة كانت تقل مع زيادة عمق القطاع الارضي ، ومن ناحية أخرى كانت قيم عناصر (النحاس، المنجنيز، النيكل، الرصاص، الزنك) أقل من (1 < C<sub>f</sub>) وبالتالي تقييم التربة علي انها غير ملوثة بهذه العناصر، بينما كانت قيم عناصر ( الكاديوم و الحديد) تقع ما بين (3 < C<sub>f</sub> < 1) وبالتالي تقييم التربة علي انها معتدلة التلوث بهذه العناصر. بالنسبة لدرجة تلوث التربة (C<sub>d</sub>) أوضحت النتائج أن جميع قيم العناصر تحت الدراسة تقل مع زيادة استخدام مياه الصرف الصحي في الري وكذلك بزيادة عمق القطاع الارضي ، كما كانت جميع القيم (C<sub>d</sub> < 1.5) وبالتالي تقييم التربة علي انها عديمة او قليلة في درجة التلوث لجميع العناصر تحت الدراسة.

