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Soil Heavy Metals Pollution: Indexing Approach Assessment and Spatial Distribution (Assanahrah, El-Beheira Governorate, Egypt)

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AGRICULTURAL soils are receiving a tremendous amount of pollutants that lead to land degradation. Therefore, it is an urgent requirement to determine and mapping the soil heavy metals content, that is the first task of soil remediation. Assanahrah area that locates at the north part of El-Beheira governorate (North of Egypt) is surrounding by many industrial activities. Therefore, it was chosen to be the pilot area to study heavy metals soil pollution.The heavy metal concentrations (Cd, Cr, Ni, Pb, and Zn) were determined and the values for the Single Pollution Index (PI_S), Nemerow Comprehensive Index (PI_N), Geoaccumulation Index (PI_{Geo}), and Improved Nemerow Comprehensive Index (PI_{IN}) were calculated based on their values to determine the pollution level of the study area.

The results indicated that the (PI_S) had the averaged values of 25.41 (Cd), 4.77 (Cr), 11.05 (Ni), 0.63 (Pb), and 2.65 (Zn) to indicate that the studied soil could be generally described as Cd, Cr, Ni-heavy polluted, Zn-slightly polluted, and Pb-no polluted.Soil heavy metals pollution indices ((PI_S), (PI_N), (PI_{Geo}), and (PI_{IN})) marked Cd as the most pollutant heavy metal. Cr single pollution and nemerow comprehensive indices ($PI_{S_Cr}$and PI_{N_Cr}) located the studied soil into heavy pollution class (HP). Basing on single pollution (PI_{S_Ni}), nemerow comprehensive (PI_{N_Ni}) and improved nemerow (PI_{IN_Ni}) indices, the studied soils were represented Ni-heavy pollution and heavily contaminated classes. All indices revealed that Pb could be nearly considered as non-pollutant heavy metal values. PI_{S_Zn}index located the studied soils in slight pollution class. The maps which generated by Kriging methods of PI_S aspects indicated that PI_{S_Ni}gradient had extremely biased distribution in the west direction. Contrary, Cd, Pb, and Zn had uniform spatial distribution. PI_{S_Cr}had a relatively biased distribution toward to the north and east.The status of heavy metals soil pollution can be considered as a system that can be studied by its parameters.

Keywords: Soil heavy metals; Indexing method; Pollution index; Pollution indices gradient and aspects; and Geographic Information System.

Introduction

There are many sources of soil pollution by heavy metals such as industrial areas, paints, fertilizers, disposal of heavy metals, sewage sludge, animal manures, wastewater irrigation, pesticides, coal combustion residues, spillage of petrochemicals, and other different sources (Wuana and Okieimen 2011; Santos-Frances et al. 2017).

Heavy metals contamination in agricultural soils may cause functional problems of soils, plant damages, and even harm of human health through contamination of the food chain (Sidhu 2016). Soil heavy metals can be assessed by two major approaches: referring heavy metals (HM) concentrations to the standard guidelines (regulation limits) and indexing method approach. Weissmannova and Pavlovsky (2017)

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described twenty indices of the assessment of soil pollution consist of two groups: single indices and total complex indices of pollution with relevant classes of it. They also provided the classification of pollution indices in terms of the complex assessment of soil quality.

Cai et al. (2015) described indexing as a type of aggregation of environmental monitoring that is commonly used when the objective of the assessment is the evaluation of some environmental criterion for large areas, usually with planning purposes. Heavy metal pollution index (Pl) had different symbols such as (HPI) (Abou Zakhem and Hafez 2014), (I) (Zhong et al. 2015), (MPI) (Singovszka et al. 2017), and (PI) (Sarhan et al. 2021).

GIS technology enables to build soil environment's spatial database to study the spatial distribution characteristics of soil heavy metals using the spatial analysis method provided by GIS and conduct pollution assessment on them using a variety of pollution assessment methods (Bai et al. 2011; Yang et al. 2011; Praveen et al. 2012; Lu et al. 2016). The maps of heavy metal concentration in topsoil were used to establish a spatial prediction of areas where local assessment is suggested to monitor and eventually control the potential threat from heavy metals. Most of the examined elements remain under the corresponding threshold values in most of the European Union land. However, one or more of the elements exceed the applied threshold concentration of 1.2 Mkm^2, which is 28.3% of the total surface area of the European Union. While natural backgrounds might be the reason for high concentrations on large proportion of the affected soils, ancient and recent industrial and mining areas show high concentrations (predominantly of As, Cd, Pb and Hg) as well, indicating the magnitude of anthropogenic effect on soil quality in Europe (Toth et al. 2016).

Some Egyptian soils are polluted by heavy metals, where concentrations of Fe, Mn, and Zn are moderate to high (Abd El-Samie 2000). Industrial contaminated areas of Fe, Mn, Zn, Cu, Cd, Co, Ni and Pb were investigated (Bassounyet al. 2020). Levels of Pb, Ni, Co and Cd in soils nearby Cairo-Alexandria agricultural highway were evaluated (Hashim et al. 2017). In the study of Siwa Oasis soils, total concentrations were Fe (0.50 - 3.37 $mg.kg^{-1}$), Mn (94 - 288 $mg.kg^{-1}$), Zn (37 -175 $mg.kg^{-1}$), and Cu (8 to 25 $mg.kg^{-1}$), while the available concentrations were Fe (0.4 - 5.6 $mg.kg^{-1}$), Mn (0.6 - 3.2 $mg.kg^{-1}$), Zn (0.4 - 1.6 $mg.kg^{-1}$), and Cu (0.1 - 1.1 $mg.kg^{-1}$), decreasing with soil depth (Abd El-All et al. 2003). In El-Maraqi region, the total Cu had the lowest value (0.98 - 13.59 $mg.kg^{-1}$), while it had the highest value in Aghormi region. Cr and Co had a moderately spread distribution pattern, while Cu, Ni, and Pb were characterized by a narrow-spread distribution pattern (Bahnasayawy 2006). Recently, geographical information systems have been used for assessing and mapping soil pollution with heavy metals in Egypt (Elbasiouny 2018; Ismail et al. 2019; Abdurrahman et al. 2020; El-Rawy et al. 2020; Salman et al. 2021; Abowaly et al. 2021).

The research aimed to (1) Assesses heavy metals pollution of Assanahrah area (Egypt) to support and encourage the efforts of soil remediation (2) Introduce a reliable approach to define priority protection areas by mapping gradient and aspects of the single soil pollution index (PI_S), and (3) Characterize the spatial distribution of soil heavy metals (HM) pollution of the studied area.

Materials and Methods

The study was elaborated through five stages (Fig. 1).

Building-up spatial database

Spatial database was built by elaborating the following processes: maps collection, digitizing, mosaicking, and clipping. Nine 1: 50000-scaled topographic maps (Egyptian Survey Authority 1998) were digitized using Arc-GIS 9.3 software (ESRI 2009). Then, the digitized topographic maps were merged in one map (mosaic map process) to clip the studied area. Assanahrah studied area that located at region El-Beheira governorate, covers an area of 8.323×8.323 km (Fig. 2).

Soil sampling

Twenty-five soil surface samples (0-40 cm) were collected using a systematic nested gridding soil sampling design of 5×5 samples with 1 km spacing. Each sample was positioned by Global Positioning System (GPS) to record the Universal Transverse Mercator (UTM) coordinates.

Determination of soil heavy metals contents

Samples (20 g) of dried soil were finely powdered by an agate ball-grinder and sieved to pass 0.15 mm nylon sieve, to determine the total heavy metals soil content. The powdered samples (0.2 g) were then digested by trace metal grade acids (9.0 ml of HNO3 and 3.0 ml of HF) using a MARS microwave digestion system according to EPA method 3052 (US EPA 1999). After

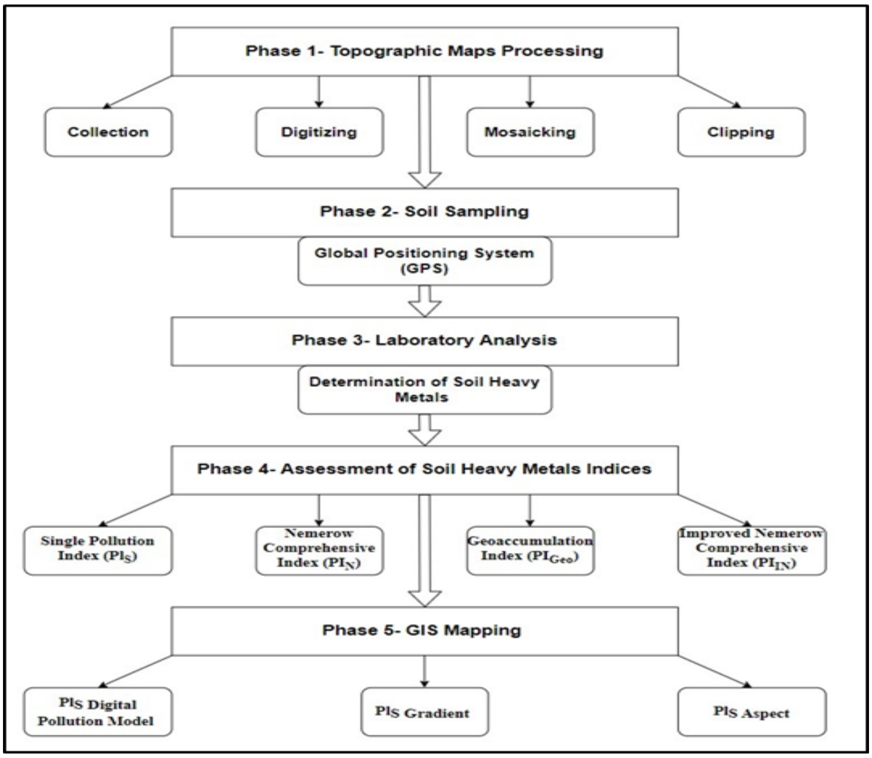

Fig. 1. Research flowchart

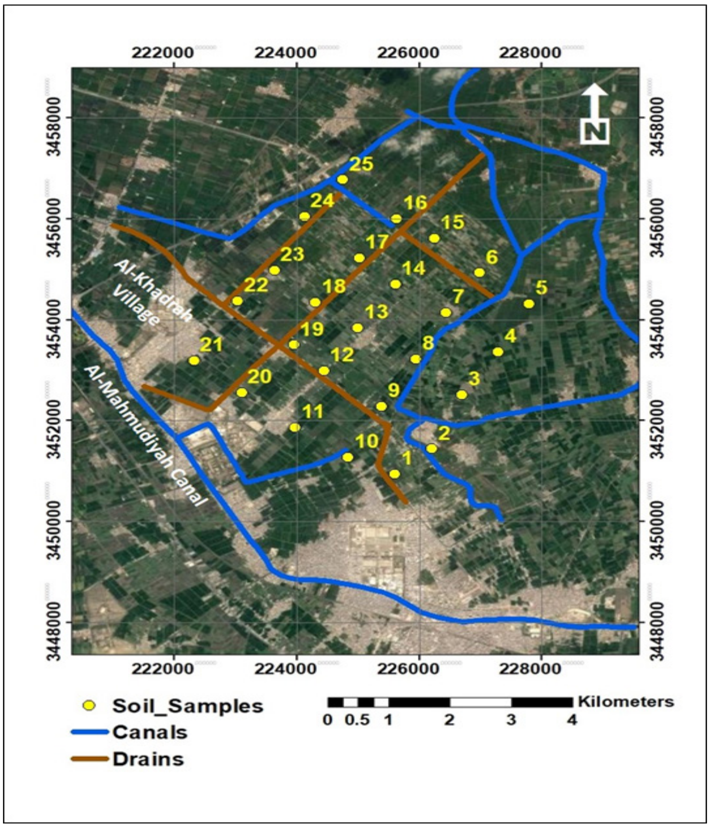

Fig. 2. Locations of soil sampling

evaporating the digestion liquids to near dryness to remove HF, the residuals were re-dissolved with dilute HNO3 and diluted with triple distilled water. A 7700 X Inductively Coupled Plasma-Mass Spectrometer (Agilent, USA) measured the concentrations of heavy metals in the final solutions. The instrument was calibrated before each set of measurements (US EPA 1999; Shahbazi and Beheshti 2019).

Assessment of soil heavy metals pollution by indexing approach

Soil heavy metals pollution was assessed by different indices; Single Pollution (Pl_S), Nemerow Comprehensive (PI_N), Geoaccumulation (PI_{Geo}), and Improved Nemerow Comprehensive (PI_{IN}).

•*Single Pollution Index (PlS)*

(Pl_S) for heavy metal (i) =Ci/Si (Nwajei *et al.* 2014).

where C_i is heavy metal concentration of a soil sample, and (Si) is its reference value. Si values of Cd, Cr, Ni, Pb, and Zn were assigned from the standards for soil environmental quality of China (Table 1) (Hu et al. 2013).

• *Nemerow Comprehensive Index (PIN)*

$$PIN=\sqrt{((PIS\ ave)^2+(PIS\ \ max)^2)/2)}$$

(Cheng et al. 2007; Hong-gui et al. 2012).

where $PI_N$is the comprehensive pollution index of the studied area (n), PI_S ave and PI_S max are the mean and maximum of the single pollution indices for each individual heavy metal, respectively.

• *Geoaccumulation Index (PIGeo)*

$PI_{Geo} = \log_2(Cn/1.5\ Bn)$ (Hong-gui et al. 2012).

where C_n is the heavy metal concentration in the soil samples, and B_n is the geochemical background value in the average shale of the heavy metal element. The constant 1.5 compensates for the natural fluctuations of a given metal and for minor anthropogenic impacts.

•*Improved Nemerow Comprehensive Index (PIIN)*

The traditional nemerow index was improved by replacing the single factor index with Igeoindex value. The following equation was applied:

$$PI_{IN\,=}\sqrt{((PIGeo\ ave)^2+(PIGeo\ max)}^2)/2) \quad \text{(Cheng et al. 2007)}$$

where PI_{IN} is the improved nemerow comprehensive index; PI_{Geo} ave and PI_{Geo} max are the mean and maximum values of the geoaccumulation index, respectively.

(Pl_S) data of the selected heavy metals (Cd, Cr, Ni, Pb, and Zn) were statistically processed to output the descriptive statistics with mean confidence intervals (CI)at the probability of 68% and 95% (Benjamini, 1988).

GIS-mapping of soil single pollution index (PlS)

The georeferenced data of the single soil pollution index (PI_S) were processed to map of HM soil pollution by GIS software Arc GIS 9.3) (ESRI 2009) using kriging method for interpolation. An innovative approach was introduced by digital pollution model (DPM). This model is so like to the well-known model of the Digital Elevation Model (DEM). The unique and main difference between DPM and DEM is the substitution of the elevation data by soil single pollution index values. DPM that presented the two-dimensional classes HM pollution enabled to map of PI_Sgradient (PI_S rate of change %) and PI_Saspects (PI_S direction of change). Directions of PI_S change was described according to their Azimuth ranges (Table 2) (FAO 1990). Finally, the spatial distributions of PI_S values, gradient %, and aspects were described by uniform, dispersed, and cluster (clumped) terms.

TABLE 1. Geochemical background value (mg/kg) of heavy metals (Hong-gui et al. 2012)

Metals	**As**	**Cd**	**Cr**	**Cu**	**Pb**	**Ni**	**Zn**	**Hg**
Values (si)	12.70	0.10	67.30	22.50	21.00	31.00	65.40	0.02

TABLE 2. Aspect class and their Azimuth range (FAO 1990)

Aspect (class)	**Compass Direction**	**Azimuth Range (Degree)**
1	North	0.00° – 22.5° and 337.5° – 360.0°
2	Northeast	22.5° – 67.5°
3	East	67.5° – 112.5°
4	Southeast	157.5° – 112.5°
5	South	202.5° – 157.5°
6	Southwest	247.5° – 202.5°
7	West	247.5° – 292.5°
8	Northwest	337.5° – 292.5°

Results and Discussion

Assessment of Soil Heavy Metals Pollution by Indexing Approach

The indices values of soil heavy metals pollution were calculated to (1) design a general view of the pollution by the selected heavy metals; Cr, Cd, Ni, Pb, and Zn, (2) assess the pollution level by basing on the standard tables, (3) elaborate critical comparison among the indices of soil heavy metals pollution, and (4) study the spatial distribution of soil single pollution index.

Assessment of soil heavy metals pollution by single pollution index (PlS)

The single pollution index (PIS) for the soil samples were calculated (Table 3) to determine the different pollution classes, according to (PIS) evaluation grading standards (Table 4). These standard thresholds of PI_S pollution classification were applied to determine the different pollution classes (Table 3). The table indicated that all soil samples located in the class of high pollution (HP) for Cd single pollution index ($PI_{S\,Cd}$). This generalization excluded samples n. 4 and that had a very low single pollution index to be classified as clean (C) class. The descriptive statistics of Cd single pollution index ($PI_{S\,Cd}$) showed that it ranged between 0.10 (sample n. 4) and 70.82 (sample n. 25) (Table 5). This expressed the wide range of Cd soil pollution that had standard deviation of value of 16.71. Cd single pollution index ($PI_{S\,Cd}$) had a mean value 25.41, while mean confidence intervals (CI) were 22.07 - 28.75 (at probability 68%) and 18.73 - 32.09 (at probability 95%), respectively (Table 5).

Cr single pollution index (PI_{S_Cr}) had moderate values with a mean of 4.77. It ranged from a minimal of 1.39 (sample n. 1) to a maximal of 12.55 (sample n.20) (Table 3). The Cr single pollution index ($PI_{S\,Cr}$) descriptive statistics specified the mean confidence intervals 68% and 95% by the ranges of 4.30-5.24 and 3.83-5.71, respectively, (Table 5). The referred that soil samples had Cr single pollution index ($PI_{S\,Cr}$) value greater than 3 to be designed heavy pollution (HP).

Ni single pollution index ($Pl_{S\,Ni}$) ranged between 0.01 (sample n. 7), and 30.43 (sample n.18) with an averaged value of 11.05. The high difference between the minimum and maximum value was reflected by high $Pl_{S\,Ni}$ standard deviation (9.19), (Table 5). Ni single pollution index ($Pl_{S\,Ni}$) values designed the majority of soil samples as heavy pollution grade, where 22 samples were located into this class. The other pollution classes were presented by sample n. 7 (clean), sample n. 1 (potential pollution), and sample n. 16 (slight pollution).

Pb single pollution index ($Pl_{S\,Pb}$) had a low average (0.63) to refer that the studied area is generally free from Pb pollution. All soil samples had values less than one to be designed as clean locations. Only sample n. 23 had Pb single pollution index value of 8.78 to be classified as heavy pollution. The mean confidence intervals 68% and 95% had the ranges of 0.29 - 0.98 and -0.07 - 1.33, respectively (Table 5). The lower limit of Pb single pollution index mean confidence intervals 95% had a negative sign (-) to reveal that some soil locations were a far way to be polluted by Pb.

Zn single pollution index ($Pl_{S\,Zn}$) values were widely distributed so soil samples located in all pollution classes; clean class (sample n.1), potential pollution (samples 2 ,4, 5, 10, 14, 15, and 16), slight pollution (samples 3, 6, 7, 8, 23, 24, and 25), and heavy pollution (samples 9, 12, 17, 18, 19, 20, 21, and 22). The mean confidence intervals (CI) 68% and 95% had the ranges 2.66-2.93, and 2.09-3.21 (Table 5). These high (CIs) indicated that most soil samples might locate in the classes of slight pollution (SP) and heavy pollution (HP).

Concisely, the single pollution index (Pl_S) had the averaged values of 25.41 (Cd), 4.77 (Cr), 11.05 (Ni), 0.63 (Pb), and 2.65 (Zn) to lead to conclude that the studied soil could be described as Cd, Cr, Ni- heavy polluted, Zn- slightly polluted and Pb-no polluted.

Assessment of soil heavy metals pollution by nemerow comprehensive index(PIN)

The single index method only evaluated the pollution of five heavy metals in each soil samples. But, these results did not accurately reflect the comprehensive pollution of the studied area caused by each kind of heavy metals. So, it was necessary to assess the overall pollution of each heavy metal by applying nemerow comprehensive pollution index (PI_N), (Cheng et al. 2007). The values nemerow comprehsive indices indicated that of Cd nemerow comprehensive index ($PI_{N\,Cd}$) and Ni nemerow comprehensive index ($PI_{N—Ni}$) had higher than these of Cr, Pb and Zn, (Table 6). But all nemerow comprehensive index of the studied heavy metals had high content, and by consequence, the studied area was classified as heavy polluted one, according to Hong-gui et al. (2012) (Table 7). So the measures must be instantly taken to avoid more HM soil pollution, and remediation actions must be carried to clean up the soil from these metals.

TABLE 3. Single soil pollution index (PIS) and heavy metals pollution classes of soil samples

Soil Sample No.	Heavy Metals									
	Cd		Cr		Ni		Pb		Zn	
	PI	PC	PI	PC	PI	PC	PI	PC	PI	PC
1	12.89	HP	1.39	PP	1.931	PP	0.03	C	0.58	C
2	24.04	HP	5.13	HP	11.92	HP	0.35	C	1.48	PP
3	12.08	HP	5.84	HP	5.60	HP	0.50	C	2.59	SP
4	0.10	C	2.86	SP	3.13	HP	0.07	C	1.51	PP
5	11.50	HP	1.72	PP	3.32	HP	0.10	C	1.20	PP
6	12.35	HP	4.17	HP	1.35	PP	0.09	C	2.45	SP
7	7.26	HP	3.63	HP	0.01	C	0.25	C	2.35	SP
8	4.26	HP	3.10	HP	1.09	PP	0.01	C	2.50	SP
9	29.6	HP	3.83	HP	6.02	HP	0.01	C	3.36	HP
10	25.55	HP	2.43	HP	3.58	HP	0.10	C	1.17	PP
11	32.49	HP	6.37	HP	16.25	HP	0.26	C	2.07	SP
12	34.04	HP	8.44	HP	25.77	HP	0.78	C	4.56	HP
13	38.21	HP	3.40	HP	8.92	HP	0.15	C	2.39	SP
14	28.93	HP	3.19	HP	6.30	HP	0.24	C	1.86	PP
15	38.45	HP	4.08	HP	5.58	HP	0.30	C	1.80	PP
16	22.69	HP	3.63	HP	2.79	SP	0.28	C	1.45	PP
17	3.63	C	4.214	HP	16.21	HP	0.23	C	3.37	HP
18	26.91	HP	6.23	HP	30.43	HP	0.38	C	6.04	HP
19	18.02	HP	4.63	HP	11.02	HP	0.24	C	3.22	HP
20	29.01	HP	12.55	HP	8.55	HP	0.68	C	3.44	HP
21	48.76	HP	7.38	HP	18.84	HP	0.57	C	3.80	HP
22	15.85	HP	5.63	HP	29.93	HP	0.47	C	6.30	HP
23	51.48	HP	4.97	HP	18.14	HP	8.78	HP	2.08	SP
24	37.19	HP	5.95	HP	18.99	HP	0.40	C	2.33	SP
25	70.82	HP	4.55	HP	20.46	HP	0.59	C	2.40	SP
Mean	35.88	HP	4.78	HP	11.05	HP	0.63	C	2.652	SP

Note: C is Clean, PP is Potential Pollution, SP is Slight Pollution, and HP is Heavy Pollution.

TABLE 4. Evaluation grading standards of the single soil pollution index (PIS) (Hu et al. 2013; Nwajei et al. 2014)

PI_S Value	$PI_S<1$	$1 \le PI_S< 2$	$PI_S< 3 \ge 2$	$PI_S \ge 3$
Pollution Class	Clean (C)	Potential (Pollution (PP	Slight Pollution (SP)	Heavy Pollution (HP)

TABLE 5. Descriptive statistics of single soil pollution index (PIS) index

Statistical Parameters	Heavy Metals				
	Cd	Cr	Ni	Pb	Zn
Mean	25.41	4.77	11.05	0.63	2.65
Min	0.1	1.39	0.01	0.01	0.58
Max	70.82	12.55	30.43	8.78	6.3
STD_DEV	16.71	2.33	9.19	1.71	1.4
SE	3.34	0.47	1.84	0.35	0.28
Mean + SE	28.75	5.24	12.89	0.98	2.93
Mean - SE	22.07	4.3	9.21	0.29	2.66
(CI) 68%	22.07 - 28.75	4.30 - 5.24	9.21 - 12.89	0.29 - 0.98	2.66 - 2.93
Mean + 2 SE	32.09	5.71	14.73	1.33	3.21
Mean -2 SE	18.73	3.83	7.37	-0.07	2.09
(CI) 95%	18.73 - 32.09	3.83 - 5.71	7.37 - 14.73	-1.4	2.09 - 3.21

Note: (CI) 68% and (CI) 95%: mean confidence interval at probability 68% and 95%

TABLE 6. Nemerow comprehensive index (PIN) and heavy metals pollution classes of soil samples

Pollution Parameter	Heavy Metal (HM)				
	Cd	Cr	Ni	Pb	Zn
PI_N Value	48.12	8.66	20.74	4.71	4.48
Pollution Class			Heavy Pollution		
Pollution Order			5		

TABLE 7. Evaluation grading standards of nemerow comprehensive index (PIN) (Hong-gui et al. 2012)

PI_N Value	$PI_N \leq 0.7$	$0.7 < PI_N \leq 1$	$1 < PI_N \leq 2$	$2 < PI_N \leq 3$	$PI_N > 3$
Pollution Class	Clean	Waring Limit	Slight Pollution	Moderate Pollution	Heavy Pollution

Assessment of soil heavy metals pollution by Geoaccumulation index (PIGeo)

The traditional nemerow index uses a single factor index method as the basis of the degree of contamination. So, it couldn't accurately reflect the heavy metal contamination with the impact of human behaviors. Therefore, in this study, the geoaccumulation index could reduce the interference of parent materials and prominent artificial effects on soil contamination by using geochemical background value of heavy metals (Table 1).

Geoaccumulation index (PI_{Geo}) classified the soil samples into different pollution classes, (Table 8) (Muller 1969). This classification conduced to draw an overview of heavy metals soil pollution, (Table 9 and Table 10). Table 10 generally showed that Pb Geoaccumulation index ($PI_{Geo—Pb}$) had the lower average (-2.01) representing uncontaminated class, meanwhile the averaged Cd Geoaccumulation index ($PI_{Geo—Cd}$) arrived to 4.64 expressing heavily to extremely contaminated class. The maximum $PI_{Geo—Cd}$ was marked in the case of sample no. 20 that may exhibit highly adverse Cd effects on human health and ecological safety (Guan et al. 2014). The PI_{Geo} highest values of Cd referred that it was the most pollutant heavy metal, where 32% and 52% were Cd heavily to extremely contaminated and extremely contaminated, respectively (Table 11). As a catalyst and an intermediate product, Cd is widely used in electroplating, chemical, electronics, non-ferrous metals, and nuclear industries. These industries are main sources of Cd contamination. Contrary, 96% of samples were categorized as Pb uncontaminated class.

Cr has been widely recognized as a heavy metal that causes serious harm to human health and is known to have carcinogenic and teratogenic effects. In the case study, the averaged value of Cr Geoaccumulation index (PI_{Geo_Cr}) was 1.94 to showed that the overall level of Cr contamination is moderately contaminated. The PI_{Geo_Cr} index ranged from 0.31 to 3.48. Consequently, Cr contamination extended from class 2 (uncontaminated to moderately contaminated) to class 4 (heavily contaminated classes) (Table 10).

The distribution of the studied soil samples basing on Ni Geoaccumulation index (PI_{Geo_Ni}) was so scattered that they spread all over six pollution classes: (12%) uncontaminated, (4%) uncontaminated to moderately contaminated, (16%) moderately contaminated, (24%) moderately to heavily contaminated, (20%) heavily contaminated, and (24%) heavily to extremely contaminated classes (Table 11). The results of the present work showed great differences between Pb Geoaccumulation index (PI_{Geo_Pb}) and Zn Geoaccumulation index (PI_{Geo_Zn}).The averaged PI_{Geo_Pb} was only -2.01 to indicate that the soil samples are so far to be nearly Pb contaminated. By contrast, the highest PI_{Geo_Zn} value of was 2.68, and its average was 1.25 (moderately contaminated) (Table 10). But it is worth to referee that Zn has been extensively documented as one of the most readily mobile and concerning elements particularly because of its toxic and carcinogenic effects.

Assessment of soil heavy metals pollution by improved nemerow comprehensive index (PIIN)

As PIGeo could reduce the effects of parent rocks and prominent artificial effects on soil heavy metal contamination, it is suitable for the evaluation of soil heavy metal contamination in industrial and mining gathering areas. However, the evaluation of PI_{Geo} is only for a single heavy metal contaminant, thus this index cannot provide a comprehensive description of the contamination status of the study area. Accordingly, an evaluation based on the comprehensive index method is necessary. The traditional nemerow index was improved by replacing the single factor index with Geoaccumulation index.

Improved nemerow comprehensive index (PI_{IN}) classification was based on the results proposed by Forstner et al. (1990) (Table 12). The PI_{IN} was calculated to assess the soil heavy metal contamination of the studied soil (Table 13). The table indicated that PI_{IN} of all soil samples ranged extended from 2.09 (Zn-uncontaminated to moderately contaminated) to 6.91 (Cd-extremely contaminated). This finding revealed serious (Cd) contamination. Cr improved nemerow comprehensive index ($PI_{IN—Cr}$) and Niimproved nemerow comprehensive index ($PI_{IN—Ni}$) considered that the studied soil as moderately to heavily contaminated and heavily contaminated ones.

Critical Comparison

Calculated indices pollution were presented by their average and pollution class (Table 13). The table formulated serious question that is there a great difference among the results of these indices?

TABLE 8. Evaluation grading standards of geoaccumulation index (PIGeo) (Muller 1969)

Class N.	PI_{Geo}	Pollution Class
0	$PI_{Geo} \leq 0$	Uncontaminated
1	$0 < PI_{Geo} \leq 1$	Uncontaminated to Moderately Contaminated
2	$1 < PI_{Geo} \leq 2$	Moderately Contaminated
3	$2 < PI_{Geo} \leq 3$	Moderately to Heavily Contaminated
4	$3 < PI_{Geo} \leq 4$	Heavily Contaminated
5	$4 < PI_{Geo} \leq 5$	Heavily to Extremely Contaminated
6	$PI_{Geo} > 5$	Extremely Contaminated

TABLE 9.Geoaccumulation index(PIGeo) and heavy metals pollution classes of soil samples

Sample N.	Cd		Cr		Ni		Pb		Zn	
	PI_{Geo}	Class N.	PI_{Geo}	Class N.	PI_{Geo}	Class N.	PI_{Geo}	Class N.	PI_{Geo}	Class N.
1	4.1	5	0.31	1	0.73	1	-4.81	0	-0.76	0
2	5	5	2.19	3	3.36	4	-1.35	0	0.59	1
3	4.01	5	2.38	3	2.27	3	-0.85	0	1.4	2
4	-3.14	0	1.35	2	1.43	2	-3.68	0	0.61	1
5	3.94	5	0.62	1	1.51	2	-3.11	0	0.29	1
6	4.04	5	1.89	2	0.22	0	-3.29	0	1.32	2
7	3.28	4	1.69	2	-6.72	0	-1.84	0	1.26	2
8	2.51	3	1.47	2	-0.09	0	-6.07	0	1.35	2
9	5.3	6	1.77	2	2.37	3	-6.62	0	1.78	2
10	5.09	6	1.12	2	1.62	2	-3.22	0	0.25	1
11	5.44	6	2.51	3	3.8	4	-1.77	0	1.08	2
12	5.5	6	2.91	3	4.47	5	-0.21	0	2.22	3
13	5.67	6	1.6	2	2.94	3	-2.6	0	1.29	2
14	5.27	6	1.51	2	2.44	3	-1.89	0	0.92	1
15	5.68	6	1.86	2	2.26	3	-1.6	0	0.87	1
16	4.92	5	1.69	2	1.26	2	-1.67	0	0.56	1
17	2.28	3	1.91	2	3.8	4	-1.97	0	1.78	2
18	5.17	6	2.47	3	4.71	5	-1.26	0	2.62	3
19	4.59	5	2.04	3	3.25	4	-1.88	0	1.72	2
20	8.59	6	3.48	4	2.88	3	-0.4	0	1.81	2
21	6.02	6	2.72	3	4.02	5	-0.66	0	1.96	2
22	4.4	5	2.33	3	4.69	5	-0.95	0	2.68	3
23	6.1	6	2.15	3	3.96	4	3.29	5	1.08	2
24	5.63	6	2.41	3	4.03	5	-1.18	0	1.25	2
25	6.56	6	2.02	3	4.14	5	-0.6	0	1.29	2

TABLE 10. Descriptive statistics of geoaccumulation index (PIGeo) and pollution classes

Statistical Parameter	Cd		Cr		Ni		Pb		Zn	
	PI_{Geo}	Class N.	PI_{Geo}	Class N.	PI_{Geo}	Class N.	PI_{Geo}	Class N.	PI_{Geo}	Class N.
Average	4.64	5	1.94	2	2.37	3	-2.01	0	1.25	2
Min	-3.14	0	0.31	0	-6.72	0	-6.62	0	-0.76	0
Max	8.59	6	3.48	4	4.71	5	3.29	4	2.68	3

TABLE 11. Frequency tables of heavy metals soil pollution classes based on geoaccumulation index (PIGeo)

Pollution Class	Class (%)				
	Cd	Cr	Ni	Pb	Zn
0	4	8	12	96	4
1	0.0	44	4	0.0	28
2	0.0	44	16	0.0	56
3	8	4	24	0.0	12
4	4	0.0	20	0.0	0.0
5	32	0.0	24	4	0.0
6	52	0.0	0.0	0.0	0.0

TABLE 12. Evaluation grading standards of improved nemerow comprehensive index (PIIN) (Forstner et al. 1990)

Class N.	PI_{IN} Values	Pollution Class
0	$0 < PI_{IN} \leq 0.5$	Uncontaminated
1	$0.5 < PI_{IN} \leq 1$	Uncontaminated to Moderately Contaminated
2	$1 < PI_{IN} \leq 2$	Moderately Contaminated
3	$2 < PI_{IN} \leq 3$	Moderately to Heavily Contaminated
4	$3 < PI_{IN} \leq 4$	Heavily Contaminated
5	$4 < PI_{IN} \leq 5$	Heavily to Extremely Contaminated
6	$PI_{IN} > 5$	Extremely Contaminated

TABLE 13. Classes and orders Soil heavy metals pollution (basing on PIS, PIN, PIGeo and PIIN indices)

Pollution Parameter	Heavy Metal (HM)				
	Cd	Cr	Ni	Pb	Zn
	1. Single Pollution Index (PI_S)				
PI_S Average	25.41	4.77	11.04	0.63	2.65
Pollution Class	Heavy Pollution		Clean	Slight Pollution	
Pollution Order	4		1	3	
	2. Nemerow Comprehensive Index (PI_N)				
PI_N Value	48.12	8.66	20.74	4.71	4.48
Pollution Class	Heavy Pollution				
Pollution Order	5				
	3. Geoaccumulation Index (PI_{Geo})				
PI_{Geo} Average	4.64	1.94	2.37	-2.01	1.25
Pollution Class	Extremely Contaminated	Moderately Contaminated	Heavily Contaminated	Uncontaminated	Moderately Contaminated
Pollution Order	5	2	3	0	2
	4. Improved Nemerow Index (PI_{IN})				
PI_{IN} Value	6.91	2.82	3.76	2.72	2.09
Pollution Class	Extremely Contaminated	Moderately To Heavily Contaminated	Heavily Contaminated	Uncontaminated To Moderately Contaminated	
Pollution Order	6	3	4	2	

Cd: The table clearly indicated that all pollution indices designed Cd as the most pollutant heavy metal. Where, the averaged of Cd single pollution index (PI_{S_Cd}) and Cd nemerow comprehensive index (PI_{N_Cd}) were 25.41 and 48.12, respectively, to locate the studied soil samples in heavy pollution class. In the case of Cd, the averaged value of geoaccumulation index (PI_{Geo_Cd}) and value of improved nemerow index ($PI_{IN—Cd}$) were 4.64 and 6.91 to categorized Cd as heavily to extremely contaminated and extremely contaminated pollution classes, respectively (Table 13).

Cr: The application of Cr pollution indices assembled soil samples into two groups. The first group consisted of the single pollution index (PI_S) that had an averaged value of 4.77, and nemerow comprehensive index value of 6.71 to consider Cr as serious pollutant heavy metals. In the meantime, the indices of the second group that formed from geoaccumulation (1.94) and improved nemerow(2.82) indices classified the studied soil as moderately contaminated and moderately to heavily contaminated ones.

Ni: With exclusion of geoaccumulation index, all other indices referred that Ni could be expressed heavy pollution case. Ni pollution indices had the averaged values of 11.05 (PI_S), values of 20.74 (PI_N), and 3.76 (PI_{IN}). These values classified Ni pollution as heavy pollution and heavily contaminated classes. Meanwhile geoaccumulation indexhad value of 2.37 to consider that Ni as moderately to heavily contaminated class (Table 13).

Pb: With exception of improved nemerow index (PI_{IN}), all indices revealed that Pb could be nearly considered as non-pollutant heavy metal values. The averaged values of Pb pollution indices were 0.63 (PI_S), value of 4.71 (PI_N), averaged value of 1.25 (PI_{Geo}) and value of 2.72 (PI_{IN}). These values located Pb into the clean, heavy pollution, moderately contaminated and uncontaminated to moderately contaminated pollution classes.

Zn: Tables (3, 7, 9, 13) clearly showed that the averaged values of Zn indices were 2.65 (PI_S), 1.49 (PI_N), 2.09 (PI_{Geo}), and 3.32 (PI_{IN}). These values designed the classes slight, heavy pollution, heavily to extremely contaminated and extremely contaminated to Zn pollution (Table 13).

Briefly, the results clearly indicated that all pollution indices designed Cd as the most pollutant heavy metal. PI_{S_Cr}andPI_{N_Cr} indices located the studied soil into heavy pollution class. Contrary, Cr geoaccumulation and improved nemerow indices classified the studied soil as moderately contaminated and moderately to heavily contaminated ones. Basing on PI_{S_Ni}, PI_{N_Ni} and PI_{IN_Ni} indices, the studied soil was considered Ni-heavy pollution and heavily contaminated classes, while they were described as members of the moderately to heavily contaminated class according to Ni geoaccumulation index. With exception of improved nemerow index (PI_{IN_Pb}), all indices revealed that Pb could be nearly considered as non-pollutant heavy metal values. The other indices distributed the soil samples through widespread pollution classes; clean, heavy pollution, moderately contaminated and uncontaminated to moderately contaminated pollution classes. PI_{S_Zn} located the studied soil in slight pollution class. However, they were distributed into heavy pollution, heavy to extremely contaminated and extremely contaminated regarding Zn pollution classes, according to the other indices.

GIS – Mapping of Soil Heavy Metals Pollution:

GIS technique was elaborated to map: (1) the spatial distribution of heavy metals pollution classes basing on the single index (PI_S), (2) PI_S gradient (rate change), and (3) PI_S aspects (direction of change). Spatial distribution is the study of things in terms of their physical locations. We are asking where things occur and how they relate to each other. The spatial distribution study works by selecting a variable and plotting incidents of that variable on a map (Study.com/academy2017). Thereby, the variables are the heavy metals concentration expressed by PI_S, gradient, and aspect.

GIS – Mapping of Soil Heavy Metals Pollution Classes:

Classes of Soil heavy metals pollutionwere mapped by the two-dimensi onal digital elevations model (DEM). Where the elevation values were replaced by those of the single pollution index (PI_S) to build the digital pollution model (DPM). The georeferenced data of the single soilpollution index (PI_S)were processed by the geographic information system (GIS) to produce a digital pollution model (DPM) of the selected heavy metals (Cd, Cr, Ni, Pb, and Zn) (Fig. 3).

The figure indicated that Cd heavy pollution class (HP) spread all over the studied area by occupying 93.81% of the studied area, while the rest is clean class (C), locating at the Northeast of the studied area, with a minor area of 6.19%. Most of the studied area is in Cr potential pollution class (HP) that presented 95.01% to spread all over the area. Cr slight pollution class (SP) that occupied 4.10% of the area distributed in three locations: Northeast, East, and Southeast (Table 14).

Cr heavy slight pollution class presented as two small patches in the Northeast and Southeast parts. The locations of the potential and slight pollution classes traced Cr cluster (clumped) spatial distribution pattern. By occupying 97.59% of the studied area, Ni heavy pollution class (HP) spread all over the studied area. The rest of the studied area grouped the classes clean (C), slight pollution (SP), and potential pollution (PP) at Northeast, Southeast, and North parts. Ni potential pollution class was characterized by a uniform spatial distribution pattern (Fig. 3 and Table 14).

The deducted data, from (PI_{S_Pb}) GIS-map (Fig. 3 and Table 14), indicated that the studied area was almost free from heavy metals pollution, where 97.71% of the studied area was categorized as a clean class (C). This high dominancy Pb clean class produced a non-spatial distribution map. The two dominant Zn classes of pollution were potential pollution (PP), and heavy pollution (HP) with percent of 34.07% and 62.66%, respectively (Table 14). Zn potential pollution class was found in the Northeast and South locations. The Northwest, West, and Southwest areas created Pb heavy pollution class. The East and Southern parts occupied by Zn slight and heavy classes presented an example of cluster (clumped) spatial distribution.

Briefly, the deducted data from (PI_S) GIS-maps showed obviously that the descending series of all soil pollution potentiality (expressing by heavy pollution class) was composed as follows; Cr (99.11%), Ni (98.14%), Cd (93.81%), Zn (65.04%) and Pb (2.29%), (Fig. 3 and Table 14).

These results indicated that the study area was mainly threatened by Cr, Ni, Cd, and Zn- high pollution potentiality. Additionally, the varia tion of PI_S gradient spatial distribution type may be due to soil elevation at the times of the deposition of anthropogenic inputs like supplementation of industrial wastes and water irrigation of El-Nasr Factory of painting silk fibres.

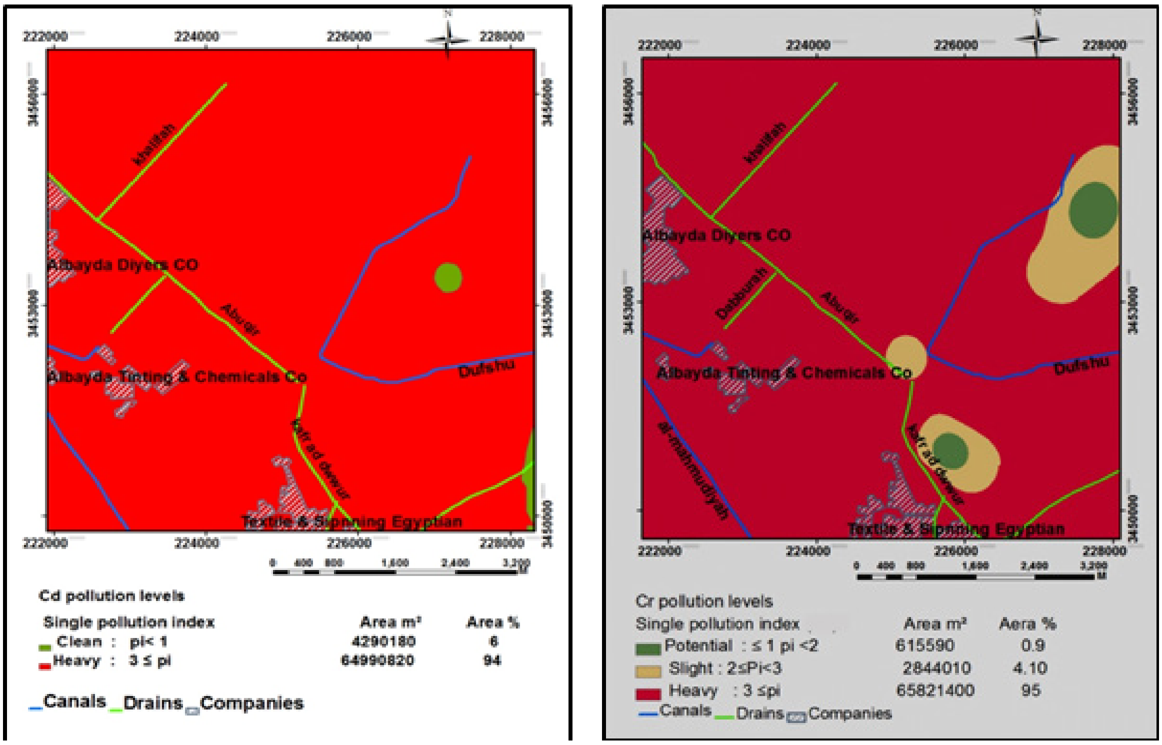

Fig. 3a. Heavy metals soil pollution classes (Cd and Cr)

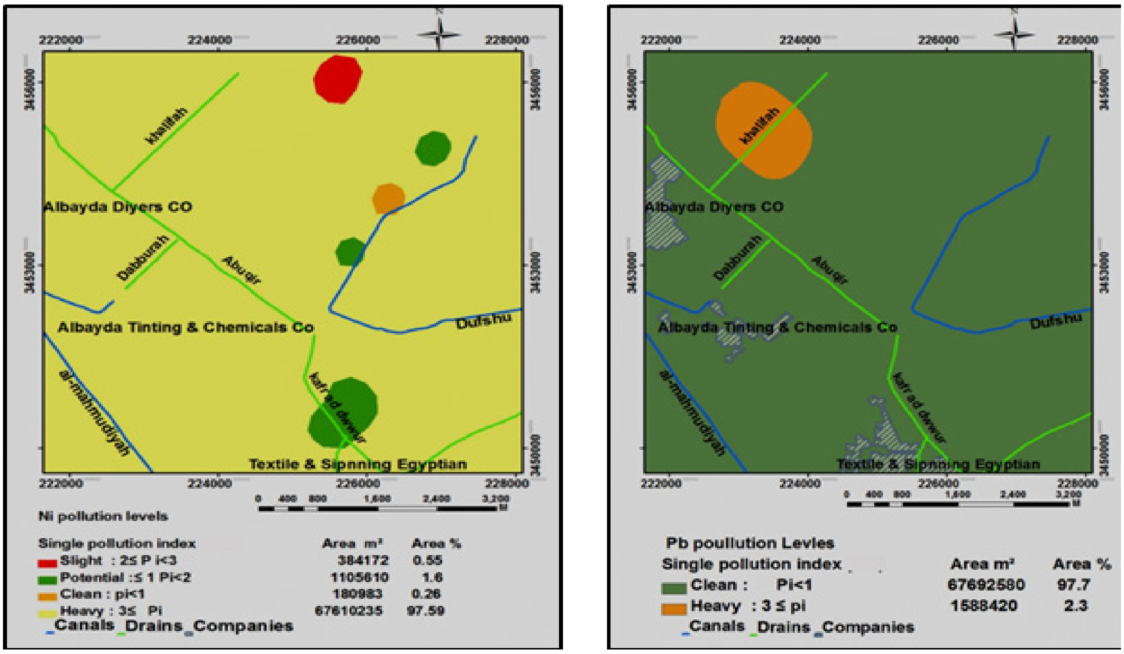

Fig. 3b. Heavy metals soil pollution classes (Ni and Pb)

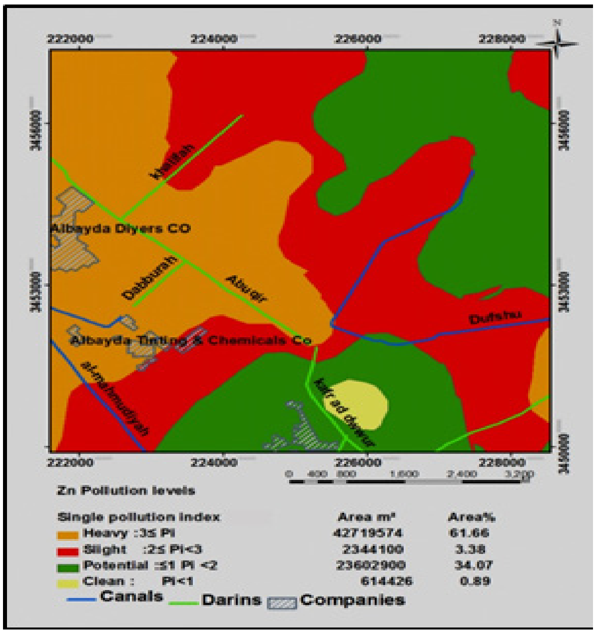

Fig. 3c. Heavy metals soil pollution classes (Zn)

TABLE 14. Spatial distribution of pollution classes (Cd, Cr, Ni, Pb, and Zn).

HM	Pollution Classes							
	Classification	Area		Location		Thresholds of (PI_S)		
		(m²)	(%)	Designation	Azimuth Range (Degree)	Min	Max	Range
Cd	C	4290180	6.19	Northeast	22.5° – 67.5°	0	≤ 1	0 - ≤ 1
	HP	64990820	93.81	Spread in all over the studied area		3	≥3	3 - ≥3
Cr	PP	65821400	95.01	Spread in all over the studied area		>1	<2	>1 - <2
	SP	2844010	4.11	Northeast East Southeast	22.5° – 67.5° 67.5° – 112.5° 112.5° – 157.5°	>1	<2	>1 - <2
	HP	615590	0.89	Northeast Southeast	22.5° – 67.5° 112.5° – 157.5°	3	≥3	3 - ≥3
Ni	C	180983	0.26	Northeast	22.5° –67.5°	0	≤ 1	0 - ≤ 1
	PP	1105610	1.60	Northeast Southeast	22.5° – 67.5° 112.5° – 157.5°	>1	<2	>1 - <2
	SP	384172	0.56	North	0.00° – 22.5°	2≤	<3	2≤ - <3
	HP	67610235	97.59	Spread in all over the studied area		3	≥3	≥3
Pb	C	67692580	97.71	Spread in all over the studied area		0	≤ 1	0 - ≤ 1
	HP	1588420	2.29	Northwest	292.5°– 37.5°	3	≥3	3 - ≥3
Zn	C	614426	0.89	South Northeast	157.5° – 02.5° 22.5° – 67.5°	0	≤ 1	0.0 - ≤ 1
	PP	23602900	34.07	Northeast South	22.5° – 67.5° 157.5° – 202.5°	>1	<2	>1 - <2
	SP	2344100	3.38	North Southeast Southwest	337.5° – 360.0° 112.5° – 157.5° 202.5° – 247.5°	≥2	<3	≥2 - <3
	HP	42719574	62.66	Northwest West Southwest	292.5° – 37.5° 247.5° – 292.5° 202.5° – 247.5°	3	≥3	3 - ≥3

Note: C is Clean, PP is Potential Pollution, SP is Slight Pollution, and HP is Heavy Pollution..

Pollution Gradient (Rate of Change (%) of Single Pollution Index (PI_S)):

Pollution gradient of single pollution index (PI_S) was graphically presented by gradient map that was derived from the two-dimensional digital pollution model (DPM) of the selected heavy metals (Fig. 4).

The figure indicated generally that PI_S values of all studied heavy metals had high change rates; PI_S gradients of Cd (98.50 %), Ni (97.64 %), Pb (96.44 %), and Cr (77.11 %), (Table 15). Contrary, Zn had a low PI_S gradient (less than 25 %) at 61.66 % of the studied area to represent the zones of low PI_{S_Zn} rate of change (Table 15).

At the Southwest parts of the studied area, the gradient of the single pollution index (PI_S) of Cd, Pb, and Zn symbolized cluster or clumped spatial distribution pattern. While it intensively flocculated spread in the gradient maps of Cr and Ni to give an example of random spatial distribution. The variation of PI_S gradient spatial distribution type may be due to slope soil variation at the times of the deposition of anthropogenic inputs like supplementation of industrial wastes and water irrigation of El-Nasr Factory of painting silk fibers.

Orientation of Pollution Gradient Direction:

Orientation of pollution gradient that represented the direction of the changes of (PI_S). PI_S aspects are so like the well-known term slope aspects of the digital elevation model (DEM). The unique and main difference is the substitution of slope aspects of elevation data by the orientation (directions) of PI_S rate changes. The GIS maps of PI_S aspects (Figure 5) indicated that PI_{S_Ni} had an extremely biased distribution 95.25% in the direction of the West (Table 16). Contrary, Cd, Pb, and Zn had uniform spatial distribution. For example, PI_{S_Cd} had the gradient directions of the North, East, South, and West. PI_{S_Cr} hada relatively biased distribution toward to the North and East by 45.15 % and 34.18 %, respectively (Figure 5 and Table 16). These different distribution patterns might conduct to refer to the variation of soil slope directions.

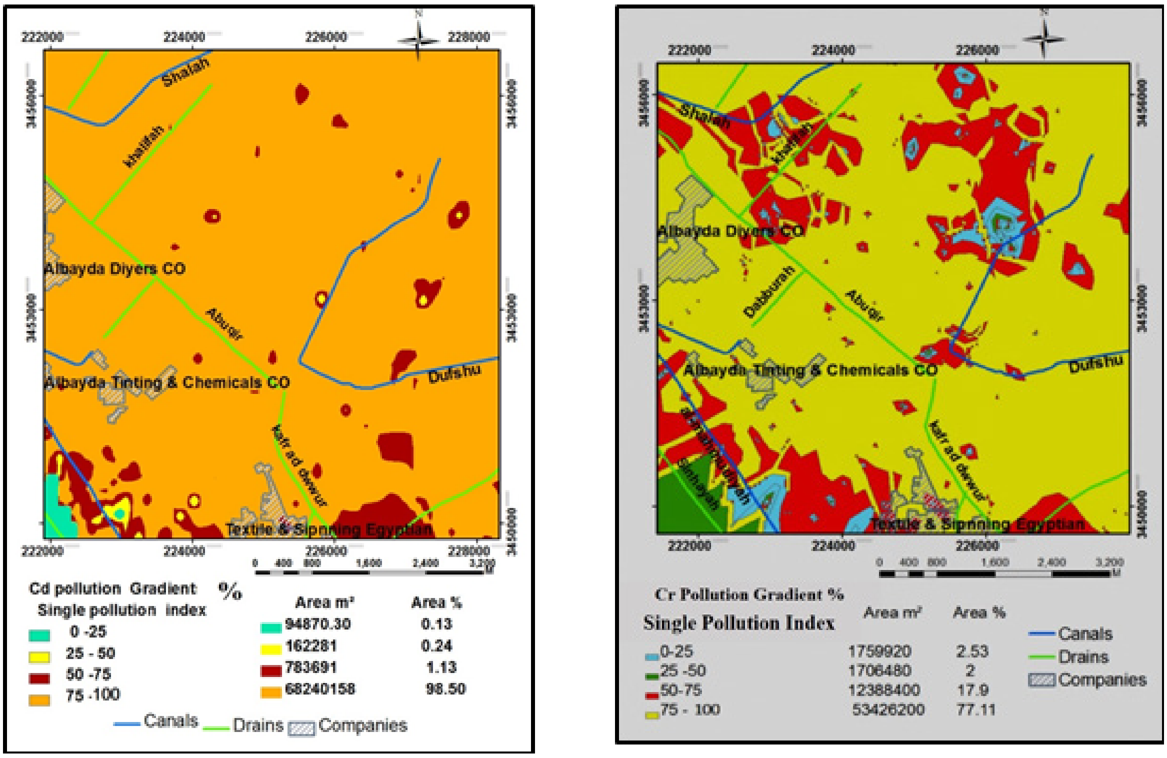

Fig. 4a. Heavy metals pollution gradient (Cd and Cr)

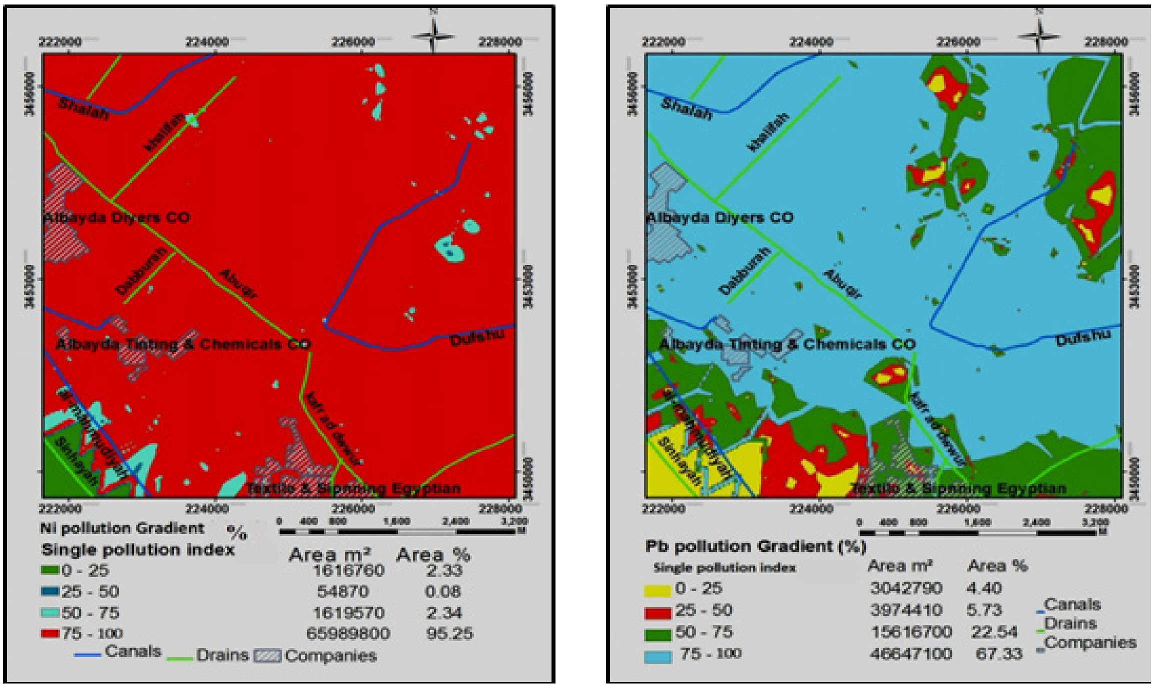

Fig. 4b. Heavy metals pollution gradient (Ni and Pb).

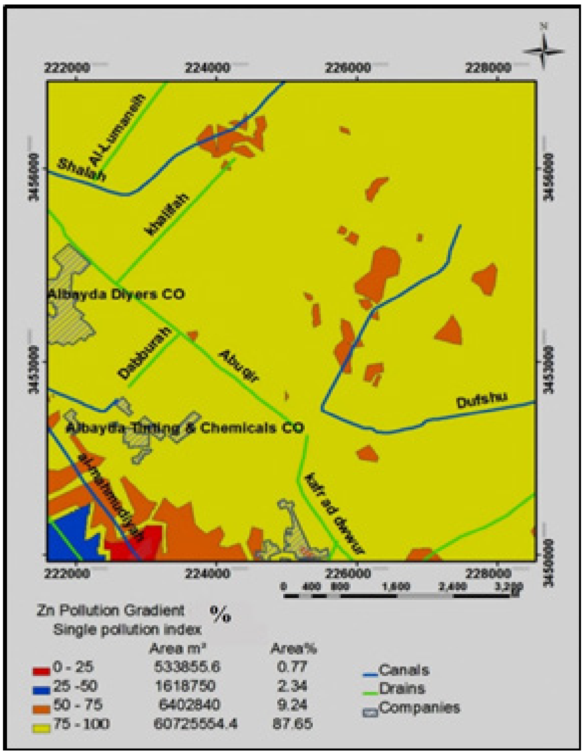

Fig. 4.c Heavy metals pollution gradient (Zn).

TABLE 15. PI_S Gradient (PI_S rate change) of Cd, Cr, Ni, Pb, and Zn heavy metals

Cd		Cr		Ni		Pb		Zn	
Gradient (%)	Area (%)	Gradient (%)	Area (%)	Gradient (%)	Area (%)	Gradient (%)	Area (%)	Gradient (%)	Area (%)
0 -25	o.13	0- 25	2.53	0 -25	2.34	0 -25	2.30	0 -25	61.66
25 - 50	0.24	25 - 50	2.00	25 - 50	0.01	25 - 50	0.05	25 - 50	3.38
50 - 75	1.13	50 - 75	17.9	50 - 75	0.01	50 - 75	1.21	50 - 75	34.07
75 - 100	98.50	75 -100	77.11	75 - 100	97.64	75 - 100	96.44	75 - 100	0,89

TABLE 16. PIS aspects (PIS directions rate change) of Cd, Cr, Ni, Pb, and Zn heavy metals

Cd		Cr		Ni		Pb		Zn	
Aspects	Area (%)	Aspects	Area (%)	Aspects	Area (%)	Aspects	Area (%)	Aspects	Area (%)
North	24.50	North	45.15	North	2.33	North	19.95	North	33.91
East	33.10	East	34.18	East	0.08	East	34.48	East	19.30
South	19.20	South	10.22	South	2.34	South	24.48	South	27.25
West	23.20	West	10.45	West	95.25	West	21.09	West	19.54

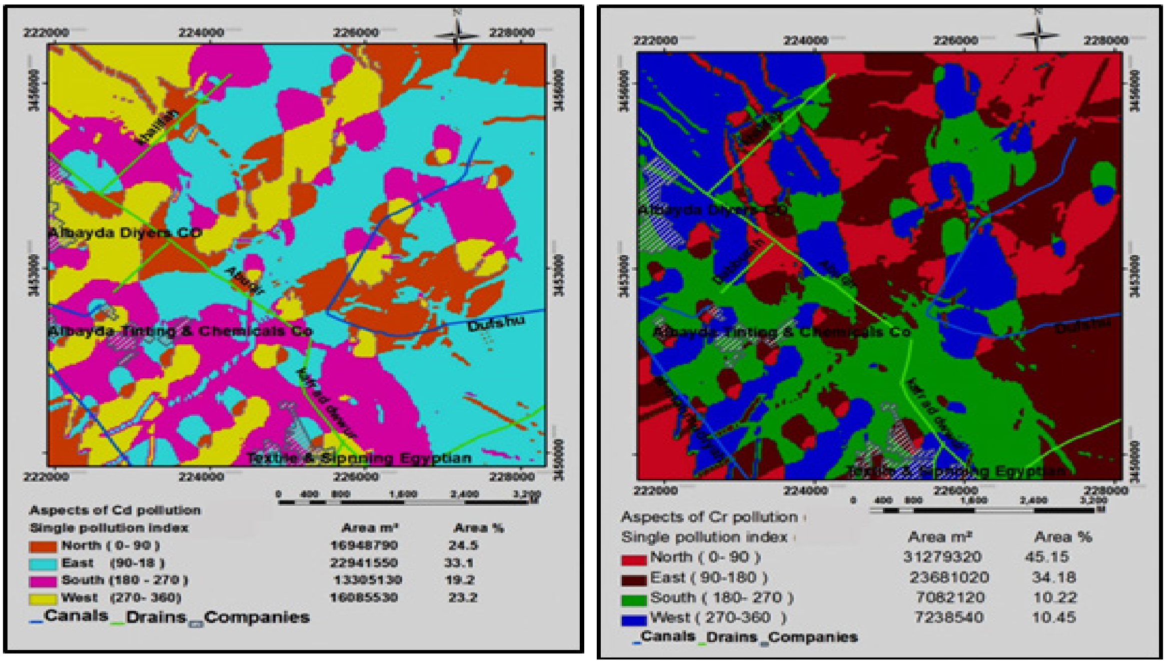

Fig. 5a. Heavy metals pollution aspects (Cd and Cr)

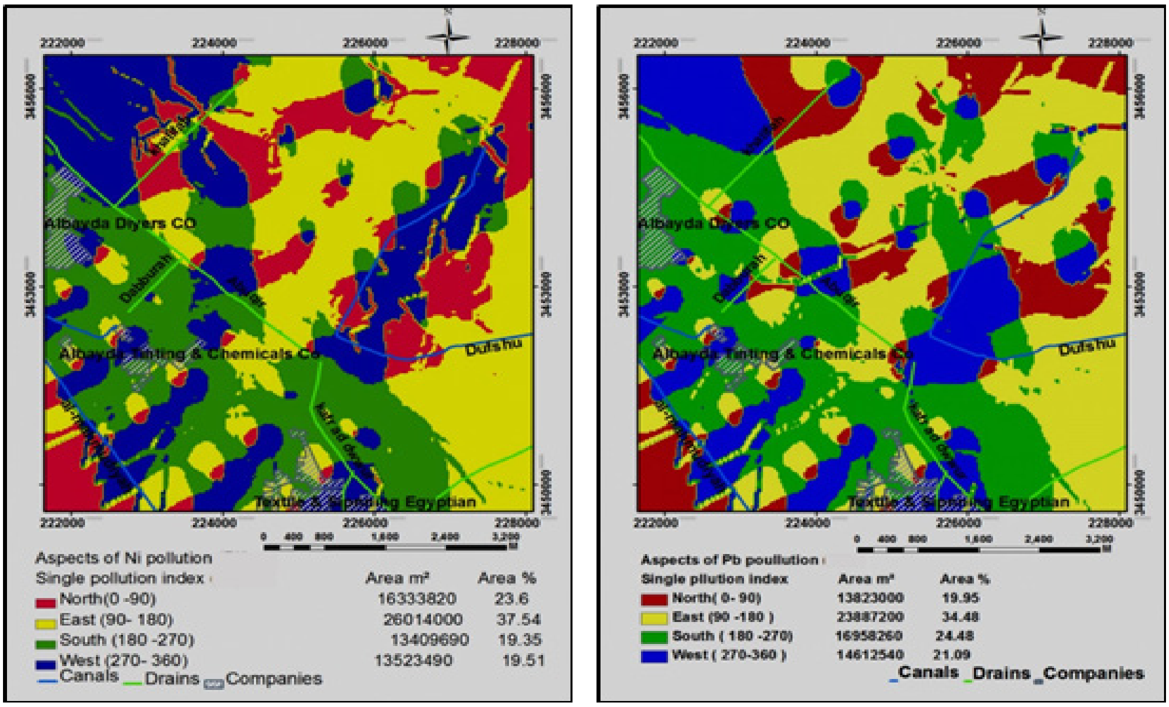

Fig. 5b. Heavy metals pollution aspects (Ni and Pb)

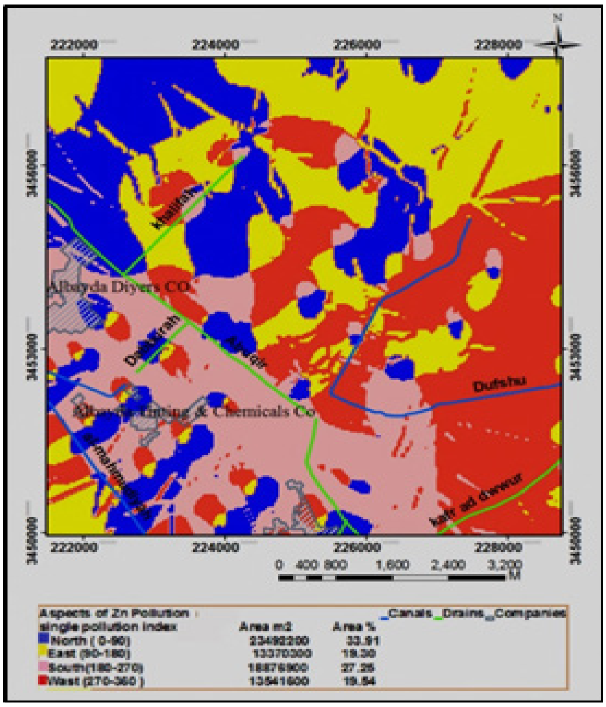

Fig. 5c. Heavy metals pollution aspects (Zn)

Conclusion

The results lead to conclude that all pollution indices designed Cd as the most pollutant heavy metal. Cr pollution categories fluctuate according to the type of single pollution and nemerow comprehensive pollution indices (PI_{S_Cr}andPI_{N_Cr})that located the studied soil into higher pollution class (HP) than that designed by the Crgeoaccumulation and improved nemerow indices. The values of PI_{S_Ni}, PI_{N_Ni} and PI_{IN_Ni} conducted to the same designation pollution to describe the studied soil as Ni-heavy pollution ones, while they were described as members of the moderately to heavily contaminated class according to Ni geoaccumulation index. With exception of Pb improved nemerow index (PI_{IN_Pb}), all indices revealed that Pb could be nearly considered as non-pollutant heavy metal values. The other indices distributed the soil samples through wide Pb spread pollution classes. The value of Zn single pollution (PI_{S_Zn}) designed the class slight to the soil, whereas the other pollution indices laid them into heavy pollution categories.

The status of heavy metals soil pollution can be considered as a system that can be studied by its parameters. So, it must first define the concerned properties to be assigned to convert into parameters. The concept of the comprehensive mapping of heavy metals soil pollution is based on the parameters: (1) determination of the heavy metals soil pollution classes to quantify the heavy metals pollution potential (2) assessment of heavy metals soil pollution tendency (gradient and aspects).

The research proved that GIS technique conducts the spatial distribution of heavy metals pollution by relating studies soil topographic features (elevation, slope gradient, and slope aspects) to heavy metals pollution parameters; pollution potential (expressed by Pl_S), rate of changes (gradient), and the direction of rate changes (aspect) of heavy metals pollution potential. Finally, this comprehensive mapping of heavy metals soil pollution classes and heavy metals spatial pollution tendency provides the capability and efficiency to compact the impact of heavy metals soil pollution.

In the case of a unique heavy metals pollution source, the characteristics of the spatial distribution of pollution may form by the soil topographic variation, elevation, slope gradient, and slope aspects. Elevation variations map the distribution and the area of pollution classes. While gradient or change rate of pollution may be attributed to the elevation gradient, and the aspect or direction of rate pollution change is determined by the soil slope aspects.

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