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Environmental modelling of heavy metals using pollution indices and multivariate techniques in the soils of Bahr El Baqar, Egypt

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Abstract Fewer studies have assessed the discharge of industrial, agricultural and municipal wastewaters on heavy metal pollution in the Soils of Bahr El Baqar, Egypt. Thirty-four samples were collected and analyzed for heavy metals, which were assessed using different indices. The summary statistics results indicate that Pb, Cd, Cu, Co, Cr and Ni concentrations in Bahr El Baqar soils are higher than those in the reference soil. By applying the Pearson rank order correlations, the result revealed that these metals have the same source of contamination. Average contamination factor values for heavy metals have an order Cd > Cr > Co > Ni > Cu > Pb > Zn > Mn > Fe, suggesting that soil samples were extremely high enriched with Cd, while Pb exhibit significant enrichment. Geoaccumulation index showed that the soils of Bahr El Baqar are having high concentrations of Cd, Ni, Co, and Cr, which exceeded the average standard value. These confirmed that Bahr El Baqar drain is facing probable environmental pollution especially with dangerous heavy metals (Pb, Cd, Co, Cr and Ni). Calculation of different indices indicates the study area falls under moderate to very high contamination degree, which regarded as polluted. These indices are useful tools for identification of anthropogenic source of soil contamination. According to this study, the agricultural activity in the Bahr El Baqar area requires careful consideration.

Keywords Enrichment factor · Contamination factor · Contamination degree · Geo accumulation index

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Introduction

The future of soil science has been changed and the priorities have been shifted from agricultural production towards environmental and ecological issues (Omran 2008). The overexploitation of resources in agriculture has led to environmental degradation: soil erosion, the greenhouse effect, and heavy metals pollution. Contamination of agricultural soils with heavy metals has always been considered a critical challenge in the scientific community. Rapid industrialization and urbanization have led to the high accumulation of heavy metals and organic pollutants in soil, water, sediment, street dust, as well as organisms in urban areas (Chaudhari et al. 2012; El Nemr 2011; Hou et al. 2013; Hu et al. 2013; Li et al. 2013; Sedky et al. 2013). Environmental contaminations by heavy metal have been increasing interest due to their toxicity and perceived persistency (Tijani et al. 2005). These metals, which are indestructible, and non-biodegradable, have toxic effects on living organisms, when permissible concentration levels are exceeded. Heavy metals frequently reported in the literature with regards to potential hazards and occurrences in contaminated soils are Cd, Cr, Pb, Zn, Fe and Cu (Akoto et al. 2008). Vehicle exhausts, as well as industrial activities emit these heavy metals so that soils, plants and even residents along roads with heavy traffic loads are subject to increasing levels of heavy metals contamination (Ghrefat and Yusuf 2006). Under certain environmental conditions, heavy metals might accumulate up to toxic concentration levels, and cause ecological damage (Bai et al. 2011; El Nemr et al. 2012).

Due to water scarcity, many countries, especially in arid and semiarid regions are forced to use low quality water in irrigation, which may be dangerous for environmental and human health. Polluted drains are considered a big threat to

the surrounded environment (Hamed et al. 2011). Bahr El-Baqar is one of the most polluted drains in Egypt (Abdel-Shafy and Aly 2002; Abdel-Azeem et al. 2007; Omran and Abd El Razek 2012). Bahr El-Baqar drain receives and carries the greatest part of the wastewater (about 3 BCM/year) into Lake Manzala through a very densely populated area of the Eastern Delta passing through Qalubyia, Sharkia, Ismailia and Port Said Governorates. The discharge of industrial, agricultural and municipal wastewaters in Bahr El Baqar drain which farmer uses it in irrigation led to contamination of these soils. These soils receive many kinds of pollutants, especially heavy metals such as lead, cadmium, nickel, and mercury, which are considered the most hazardous (Omran and Abd El Razek 2012; Park and Shin 2006). However, the question still not answers yet is "What is the environmental and ecological risk assessment of heavy metals of Bahr El Baqar on sustainable soil resources"? For an ecological risk assessment associated with pollutant exposure in ecosystems, several environmental factors must be considered, such as chemical, physico-chemical, biological, and ecotoxicological parameters. All these variables must be integrated and some indexes have been applied to do it. Varieties of methods have been developed for the risk assessment of heavy-metals as sediments enrichment factor, index of geological accumulation and pollution load index (Ohlson and Serveiss 2007; Serveiss 2002; Sun et al. 2010).

To the best of my knowledge, no reports are available on the different pollution indices to assess ecological risk of heavy metal contamination in Bahr El Baqar region. Therefore, the overall objective of the present study is to use geostatistical and multivariate analysis to assess the heavy metal contamination in some polluted soils of Bahr El Baqar region. Specific objectives of this study were to:

1. Assess the current level of heavy metal concentrations by different indices in the soils of Bahr El Baqar; and
2. Evaluate different pollution indices to assess the ecological and environmental risk due to soil contamination by heavy metal.

Materials and methods

Overall the study area and methodology

The study area is located in northern Egypt, Bahr El Baqar region, between 31°50′ to 32°20′ longitude and 30°40′ to 31°10′ latitude (Fig. 1). Environmental protection in Bahr El Baqar region is faced critical problems due to the increasing population, demolishing natural resources, environmental pollution, land-use planning as well as others (Omran and Abd El Razek 2012). Soils,

which are adjacent to El Manzala Lake, are described as heavy saline alkali low-lying clay, which is lacustrine deposits. The land surface is flat, gently sloping towards the north and ranges in elevation from below sea level to 4 m a.s.l in the highest point (Omran and Abd El Razek 2012). A reconnaissance visit was performed in the study area to get acquainted with different landscape features; land-use and land-cover patterns. A 60-km-long stretch of the Bahr El Baqar drain was selected for the present study. The extensive field surveys in August 2014 were guided with a Global Positioning System receiver. Thirty-four soil samples were collected from Bahr El Baqar region (Fig. 1). Soils were characterized by their main physicochemical properties and by their total heavy metal contents. Soil samples were collected from selected agricultural fields. Soils were randomly sampled from the upper horizon (0–20 cm) and bulked together to form one composite sample. Soil samples were air-dried and sieved through a mesh of <2 mm, and then sealed in paper envelopes until analysis. The electrical conductivity (EC) of soils was determined using 1:2 soil to solution (H_2O) ratio. Particle size distribution was determined by the pipette method (Gee and Bauder 1986). Soils were also characterized for their carbonate content (Allison and Moodie 1965). Soil pH was measured in deionized water (pH_w) and in 0.01 M $CaCl_2$ (pH_{CaCl2}) (in 1:2.5 suspensions). Organic carbon (OC) was determined by the wet digestion (Walkley and Black 1934). CEC was determined for the soil samples by Na-method (Chapman 1965). Figure 2 shows the overall methodology used in this study.

The selected indices and overall analysis

The pollution indices classified from modelling point of view into two types: single indices and integrated indices (Caeiro et al. 2005). The degree of contamination in the soil is determined with the help of following indices (Table 1): enrichment factor (EF); contamination factor (CF); contamination degree (CD); pollution load index (PLI); metal pollution index (MPI) and geo accumulation index (I-geo). Many authors prefer to express the metal contamination with respect to average shale to represent the degree of quantification of pollution. Some authors have considered the background value of their area of study (Thambavani and Mageswari 2013) to be the geometric mean of concentration of the different sample sites, which is the antilog of the arithmetic average of log10 of the concentration values. According to them, the geometric mean reduces the importance of a few high values in a sample group and therefore, is numerically less than the arithmetic mean. Such background value, however, varies from place to place. As such, this methodology of
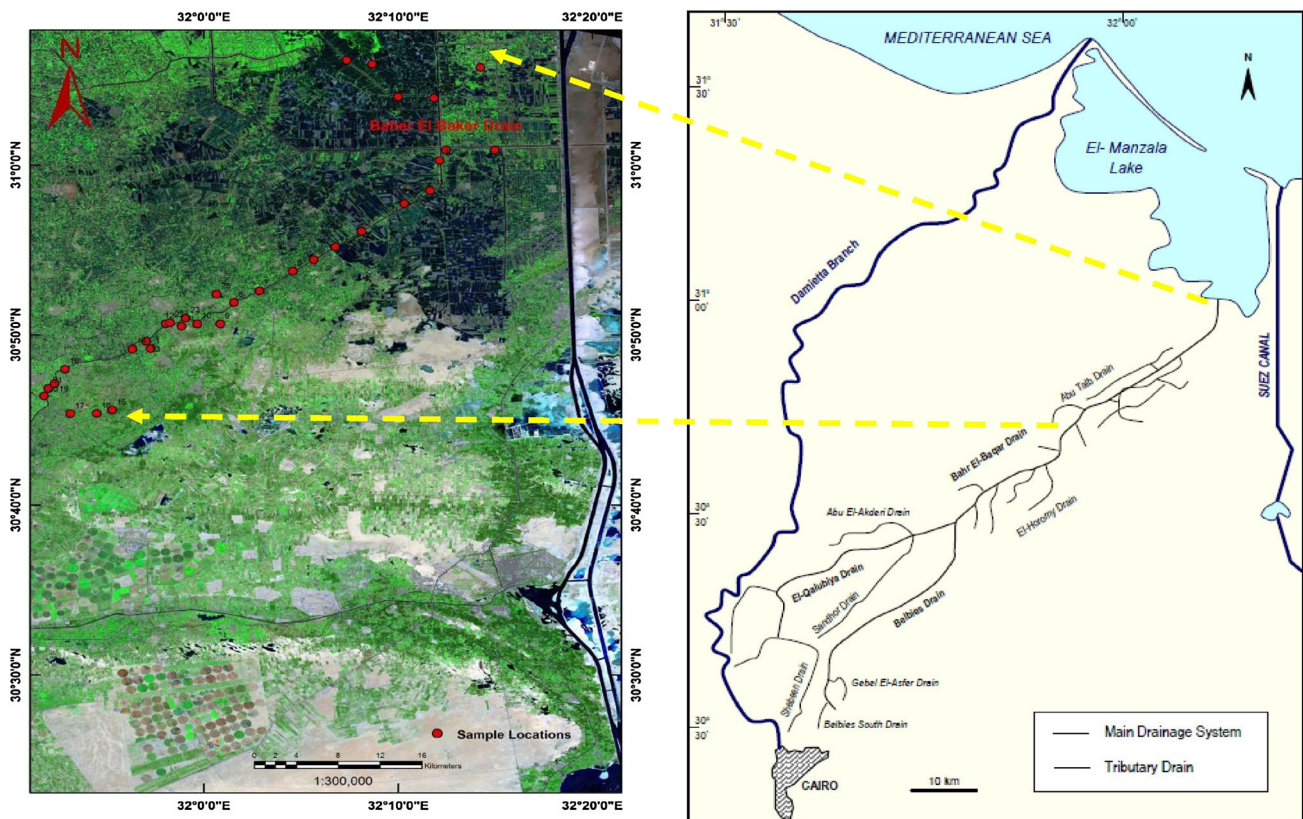
Fig. 1 Footprints and landsat-8 image for different samples locations selected for the study area

determining background value has not been considered in the present study. Instead, the world surface rock average (Omran and Abd El Razek 2012) and US EPA standard of individual metal has been taken to be the background.

## Geostatistical approach for interpolating soil heavy metals

The geostatistical approach consists of two parts: calculation of an experimental variogram and the model fitting, and estimation at unsampled locations (Burgos et al. 2006). A variogram is used to measure the spatial variability of a regionalized variable and provides the input parameters for the spatial interpolation of variogram kriging (Webster and Oliver 2001). The variogram plot is fitted with a theoretical model (spherical, exponential, linear, or Gaussian). The best fitting function can be chosen by cross-validation, which checks the compatibility between the data and the model. The goodness of fit was evaluated by the mean error (unbiasedness if its value is close to 0) and the mean squared error, which should equal the kriging variance, and the root mean square error (should be 1). The fitted model provides information about the spatial structure as well as the input parameters for kriging interpolation (Burgos et al. 2006). After selecting an appropriate variogram model, the

parameters can be used with the data to predict heavy metal concentrations at unsampled locations using kriging. Kriging is the most popular generalized linear regression techniques for minimizing and estimating variance in an unsampled location (Webster and Oliver 2001). The selection of a kriging algorithm (ordinary, simple, universal kriging, etc.) model should be guided by the characteristic of the data under study (Saito and Goovaerts 2000). Geostatistical analyses were performed using the Geostatistical analyst extension available in ESRI ArcMap v 10.2

Results and discussion

Physico-chemical properties of the Bahr El Baqar soils

The pH, CEC, clay and organic matter contents (OM) are the principal soil characteristics that determine the capacity to retain heavy metal pollutants. The average of pH in Bahr El Baqar soils is about 8.00, because of the presence of calcareous parent material (Table 2). The pH of the soil solution maintained at alkaline condition showed low mobility of all heavy metals. It could be attributed to the presence of carbonates at a high concentration. The high
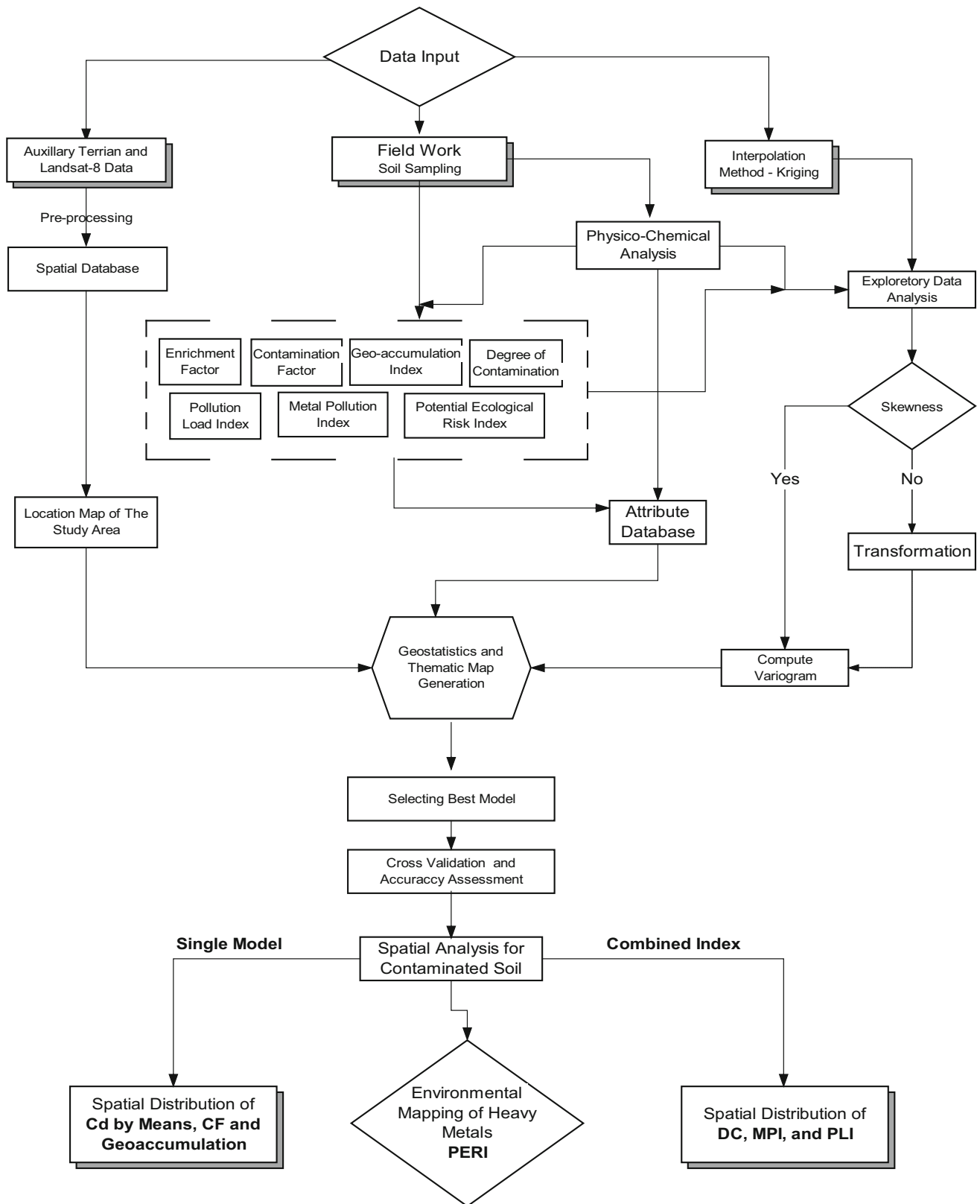
Fig. 2 Methodology and procedure used to assess the heavy metals in the soils of Bahr El Baqar, Egypt

Table 1 The different types of model, rank of the values, and categories for the description of soil contamination

Model	Equation	Description	Index/class	Contamination degree	Source
Enrichment Factor (EF)	$EF = \dfrac{(M/Fe)_{sample}}{(M/Fe)_{background}}$ EF is the enrichment factor, (M/Fe) sample is the ratio of metal and Fe concentration of the sample and (M/Fe)background is the ratio of metals and Fe concentration of a background	EF was considered to estimate the abundance of metals in samples. EF was calculated by a comparison of each tested metal concentration with that of a reference metal. The normally used reference metals are Mn, Al and Fe	EF < 2 2–5 5–20 20–40 EF > 40	Deficiency to low enrichment Moderate enrichment Significant enrichment Very high enrichment Extremely high enrichment	Rubio and Vilas (2000)
Contamination Factor (CF)	$CF = \dfrac{Csi}{Cbi}$ Csi is the measured concentration of the examined metal i in the soil sample, and Cbi is the background value of heavy metal i.	The CF is the ratio obtained by dividing the concentration of each metal in the soil by the baseline or background value (concentration in unpolluted soil)	CF < 1 1 < CF < 3 3 < CF < 6 CF > 6	Low degree of contamination Moderate degree of contamination Considerable degree of contamination Very high degree of contamination	Tippie (1984)
Geo-accumulation Index (I-geo)	$I-geo = \dfrac{Log2Csi}{1.5Cbi}$ Csi is the concentration of the element i in the samples, Cbi is the background value of the element i, and the factor 1.5 is used to take into account the possible lithological variability	The geo-accumulation index (I-geo) was used to calculate metal contamination level in the soils by comparing current concentrations with pre-industrial levels	Igeo > 5 - Class 6 4–5 - Class 5 3–4 - Class 4 2–3 - Class 3 1–2 - Class 2 0–1 - Class 1 0 - Class 0	Extremely contaminated Strongly to extremely contaminated Strongly contaminated Moderately to Strongly contaminated Moderately Contaminated Uncontaminated to moderately contaminated Uncontaminated	Sutherland (2000)
The Degree of Contamination (DC)	$DC = \Sigma\,CF$	The contamination levels may classified based on their intensities on a scale ranging from 1–6 (0 = none, 1 = none to medium, 2 = moderate, 3 = moderately to strong, 4 = strongly polluted, 5 = strong to very strong, 6 = very strong)	DC < 6 6 ≤ DC < 12 12 ≤ DC < 24 DC ≥ 24	Low Moderate Considerable Very high	Aksu et al. (1998)
Pollution Load Index (PLI)	$PLI = (CF_1 \times CF_2 \times CF_3 \times \cdots \times CF_n)^{1/n}$ n is the number of metals studied, and CF is the contamination factor	The PLI provides simple but comparative means for assessing site quality	PLI < 1 PLI = 1 PLI > 1	Perfection Base line level of pollution Deterioration of site quality	Usero and Garcia (2000)
Metal Pollution Index (MPI)	$MPI = Log \sum_{i=1}^{n5} X/refi$ refi represents a normalizer, or a reference value for each of chosen metals in selected sites, while x represents mean value of metal concentration	MPI has been calculated to enable presentation of all results from the metal concentrations as one value, yet overcoming the difficulties with both application and understanding of demanding statistical analysis	MPI < 1 MPI > 1	Non-polluted Polluted	Jorgensen and Pedersen (1994)
Potential Ecological Risk Index (PERI)	PER = TRF. CF PERI = Σ PER	The PERI provides a fast and simple quantitative value for environmental assessment	PERI < 150 150 ≤ PERI < 300 300 ≤ PERI < 600 PERI ≥ 600	Low Moderate Considerable Very high	Hakanson (1980)

Table 2 Summary statistics of soil physico-chemical properties of the Bahr El Baqar

Properties	Minimum	Maximum	Range	Mean	Std. deviation	CV	Skewness	Kurtosis
Fe	7.80	87.96	80.16	47.45	25.19	634.38	−0.04	−1.31
Mn	13.73	98.73	85.00	58.98	23.39	547.26	−0.32	−0.81
Cu	7.73	280.30	272.58	65.70	54.68	2990.45	2.27	6.69
Zn	39.67	215.58	175.91	90.56	41.86	1752.64	0.76	0.76
Ni	61.32	88.73	27.41	73.22	7.96	63.34	0.74	−0.41
Pb	24.42	52.40	27.98	36.64	7.04	49.63	0.58	0.05
Cd	10.27	19.07	8.80	14.69	2.31	5.32	0.06	−0.44
Co	70.51	113.82	43.31	89.72	12.58	158.29	0.66	−0.71
Cr	84.92	134.06	49.14	106.96	12.26	150.23	0.16	0.07
CEC	4.79	96.75	91.96	45.00	25.33	641.77	0.80	−0.50
Total N	2.38	7.70	5.32	4.58	1.28	1.63	0.71	0.33
OM	0.22	3.55	3.33	1.60	0.79	0.62	0.73	0.69
pH	7.58	8.82	1.24	8.00	0.30	0.09	0.81	0.25
$CaCO_3$	0.61	29.20	28.59	6.58	8.77	76.93	1.70	1.44
Sand	2.60	95.72	93.12	43.00	25.83	667.07	0.16	−0.78
Silt	1.04	56.89	55.85	30.38	12.93	167.09	−0.20	0.43
Clay	3.24	80.00	76.76	26.61	24.52	601.14	1.01	−0.51

Fe, Mn, Cu, Zn, Pb, Cd, Co, Cr are in mg kg^{-1}, CEC Cmolc/Kg, N mg/gm, OM, $CaCO_3$, sand, silt, clay are in %

pH value measured in soils potentially limits the risk of metal mobilization. The relatively high content of OM (1.6 %) is mainly related to the high organic matter flux to the soil due to direct discharge of domestic and industrial wastewaters. Soil organic matter is a key for sorbing phase of metals. The dissolution of humic acid at high pH is responsible for the dissolution of Cu and Pb from the soil. Organic matter is important for the retention of metals by soil solids, thus decreasing mobility and bioavailability.

The summary statistics results indicate that the mean value of soil Pb, Cu, Cr and Zn concentrations is 36.64, 65.70, 106.96, and 90.56 mg kg^{-1}, respectively and the range between the minimum and maximum values is 27.98, 272.58, 49.14, and 175.91 mg kg^{-1}, respectively, which is large. There are great variations and high skewness for Cu and Zn (2.27 and 0.76). The CV % values reflect the mean variation of each sampling site in the population. The order of the CV %s for each element, from high to low, was Cu > Zn > Fe > Mn > Co > Cr > Ni > Pb > Cd. This result showed that the variation of Cu and Zn in the soil was larger than other metals.

The mean concentration of metals at selected locations and their world surface rock average is given in Table 3. The mean value of soil Cu, Ni, Pb, Cd, Co and Cr concentrations is 65.70, 73.22, 36.64, 14.69, 89.72, and 106.96 mg kg^{-1}, respectively, which is much higher than the US EPA threshold level (16, 16, 20, 0.6, 23, and 26 mg kg^{-1}, respectively) presented in Table 3. This result shows that Pb, Cd, Cu, Co, Cr and Ni concentrations in

Bahr El Baqar soil are higher than those in the reference soil. These levels are far above the average concentrations in the earth's crust (Hasan 2007; Yobouet et al. 2010) and the threshold concentration of European Union Standards. Table 3 indicates these soils are heavily polluted with heavy metals that are part of the most dangerous industrial and municipal waste (Hasan 2007). The ranking order of mean values of the heavy metals in the Bahr El Baqar soils followed the sequence: Cr > Zn > Co > Ni > Cu > Mn > Fe > Pb > Cd.

Multivariate statistical analysis

Multivariate analysis (i.e., Principal component analysis; and correlation) has been proved to be an effective tool for providing suggestive information regarding heavy metal sources and pathways (Hu et al. 2013).

Factor analysis

Table 4 shows a factor analysis, which was, performed on raw data in an attempt to further clarify the major controlling factors that determine the heavy metal's distribution in the Bahr El Baqar soils. Four of the factors, which account for most of the variability in the 17 variables, were obtained. Four factors have been extracted which had eigenvalues greater than or equal to 1. Together they account for 78.11 % of the variability in the original data (Table 4). The types of factoring have been selected as

Table 3 Comparison of heavy metal concentrations (mg kg^{-1}) with those of the threshold and the Earth's crust

Parameters	Fe	Mn	Cu	Zn	Ni	Pb	Cd	Co	Cr
Values obtained from present study	47.45	58.98	65.70	90.56	73.22	36.64	14.69	89.72	106.96
Average concentration in the Earth's crust*	56,300	850	55	70	75	12.5	0.15	–	100
Extremes values recorded in shale**	46,700	950	40	95	68	20	0.3	–	90
Threshold concentration***EUS	–	–	140	300	75	300	3	–	150
US EPA, 1993	267.00	72.00	16.00	95	16.00	20	0.6	23	26

* Taylor 1964; ** Turekian and Wedepohl 1961; *** European Union 2002

Table 4 Factor loading component matrix after varimax rotation

	1	2	3	4
CEC	0.077	0.154	0.944	0.144
Total N	0.332	−0.500	0.377	0.086
OM	0.696	−0.171	−0.011	0.514
pH	−0.222	0.042	0.662	−0.098
CaCO$_3$	−0.078	0.067	0.812	0.393
Sand	−0.406	0.092	−0.292	0.038
Silt	0.023	−0.586	0.676	−0.271
Clay	−0.048	0.212	0.943	0.103
Fe	0.832	−0.088	0.305	−0.047
Mn	−0.058	−0.235	0.410	0.777
Cu	0.754	0.150	−0.250	−0.001
Zn	−0.021	0.311	−0.144	0.738
Ni	0.472	−0.636	0.464	0.163
Pb	0.249	0.621	−0.173	−0.578
Cd	−0.658	−0.413	−0.304	0.371
Co	−0.584	0.511	0.482	0.223
Cr	0.166	0.683	0.401	0.087
Variance %	47.244	13.652	10.718	6.492
Cumulative %	47.244	60.896	71.615	78.107

Extraction method: principal component analysis

principal components. Metal loadings of the factors have been given in Table 4. Factor1 accounts for 47.24 % of the variability in the original data. The first group of variables can be described as an anthropogenic assemblage composed of mainly Fe, Co, Cd, Cu and OM. This factor reflects the binding of heavy metals to organic matter. Factor 2 accounts for 13.65 % of the variability in the original data. The second group of anthropogenic variables composed of mainly Ni, Cr and Pb. Factor 3 accounts for 10.72 % of the variability in the original data and explains metal sorbtion pools: carbonates, alumino silicates (clay minerals), pH and CEC. The first two of them resulted from terrestrial sources. Both of them are conservative components and they lose some trace metal contents via resuspension by the winds. Factor 4 accounts for 6.49 % of the variability in the original data and is composed of mainly Mn and Zn.

Statistical analyses

By applying the Pearson (parametric) rank order correlations (Table 5), the results revealed that the Fe is well correlated with Mn and moderately correlated with Cu and Zn (r = 0.819, 0.488, 0.471 respectively). Furthermore, there is a good correlation between Ni and Co (r = 0.966). There are significant correlations between Cu and each of Zn, Pb, (r = 0.678, 0.544, respectively) as shown in Table 5. This result revealed that these metals have the same source of contamination. The results in Table 5 also showed that Ni, Fe, Mn, Cu, Zn and Co are positively correlated with OM, pH and clay, CaCO$_3$, and CEC and of course negatively correlated with sand. Clay highly correlated with Fe, Mn, Cu, and Zn (0.773, 0.658, 0.786, and 0.709 respectively).

Quantification of heavy metals accumulation

Single models

Table 6 shows the results of contamination factor (CF). As shown in Table 6, average CF values for heavy metals have an order Cd > Cr > Co > Ni > Cu > Pb > Zn > Mn > Fe, suggesting that soil samples was extremely high enrichment with Cd, while Pb exhibit significant enrichment. In contrast, the rest of the metals show moderate or minimal enrichment in the study area. With respect to specific sites, high CF values for Cd (e.g., 31.79 were found in samples 10, 18, and 23. High CF values for Cr (5.16) were found in sample 5. High CF values for Co (4.95) were found in samples 6 and 19. However, for Cu, most of the locations have moderate to considerable contamination except samples of 26 and 27 which very highly contaminated. These locations were located at the downstream and continuously receive a vast amount of wastewater and other wastes of the city. Results in Table 6 show the CF values of most of the Mn, Zn, and Fe metals in the study area, which are low degree of contamination. Nevertheless, CF values for metals like Ni, Cd and Cr shows considerable degree. This is due to the influence of external discrete sources like industrial activities,

Table 5 Pearson (parametric) rank order correlations

	CEC	Total N	OM	pH	CaCO$_3$	Sand	Silt	Clay	Fe	Mn	Cu	Zn	Ni	Pb	Cd	Co	Cr
CEC	1																
Total N	0.209	1															
OM	0.677**	0.458**	1														
pH	0.596**	0.131	0.220	1													
CaCO$_3$	0.806**	0.241	0.714**	0.580**	1												
Sand	−0.901**	−0.349*	−0.565**	−0.524**	−0.672**	1											
Silt	−0.031	0.478**	−0.066	−0.150	−0.217	−0.349*	1										
Clay	0.966**	0.115	0.630**	0.631**	0.822**	−0.869**	−0.160	1									
Fe	0.776**	0.312	0.522**	0.540**	0.652**	−0.864**	0.259	0.773**	1								
Mn	0.715**	0.455**	0.508**	0.435*	0.560**	−0.828**	0.406*	0.658**	0.819**	1							
Cu	0.769**	0.080	0.456**	0.490**	0.502**	−0.666**	−0.161	0.786**	0.488**	0.401*	1						
Zn	0.686**	0.015	0.438**	0.448**	0.532**	−0.592**	−0.160	0.709**	0.471**	0.434**	0.678**	1					
Ni	−0.439**	−0.332	−0.486**	−0.458**	−0.468**	0.458**	−0.029	−0.467**	−0.385*	−0.414	−0.429*	−0.460**	1				
Pb	0.521**	−0.014	0.226	0.392*	0.329	−0.482**	−0.147	0.586**	0.479**	0.388*	0.544**	0.467**	−0.399*	1			
Cd	−0.420*	−0.095	−0.100	−0.219	−0.297	0.344*	0.117	−0.424*	−0.337	−0.265	−0.188	−0.363*	−0.002	−0.582**	1		
Co	−0.370*	−0.330	−0.460**	−0.413*	−0.366*	0.406*	−0.060	−0.396*	−0.341*	−0.381*	−0.415*	−0.434**	0.966**	−0.420*	−0.056	1	
Cr	0.241	−0.128	0.117	0.015	0.110	−0.141	−0.099	0.201	0.092	0.118	0.076	0.449**	0.263	0.085	−0.536**	0.300	1

* Correlation is significant at the 0.05 level, ** Correlation is significant at the 0.01 level

Table 6 Contamination factor (CF) for heavy metal accumulation in the study area

Sampling point	Fe	Mn	Cu	Zn	Ni	Pb	Cd	Co	Cr
P1	0.18	0.90	2.11	0.91	4.32	1.61	25.81	3.41	4.06
P2	0.17	0.96	3.03	0.64	4.90	1.22	28.06	4.38	4.69
P3	0.20	0.97	2.94	0.43	5.44	1.70	22.25	4.80	4.17
P4	0.13	0.72	2.52	0.42	4.40	1.73	22.47	3.85	3.55
P5	0.06	0.76	5.39	1.60	4.41	1.59	21.58	3.94	5.16
P6	0.27	0.90	2.04	1.05	5.55	1.53	26.98	4.95	4.21
P7	0.27	1.11	2.62	0.60	4.62	2.04	25.36	3.83	4.14
P8	0.03	0.19	1.24	0.43	5.44	1.70	22.25	4.80	4.17
P9	0.03	0.37	0.48	1.08	4.57	2.05	26.19	3.84	4.13
P10	0.05	0.91	1.87	0.44	4.26	1.46	31.79	3.66	3.27
P11	0.06	0.75	2.20	0.91	4.36	1.65	26.14	3.42	4.04
P12	0.14	1.06	2.74	0.63	4.46	1.94	27.69	3.57	3.70
P13	0.20	0.89	6.14	1.48	4.14	2.62	19.25	3.48	4.56
P14	0.15	0.82	2.17	0.91	4.32	1.61	25.81	3.41	4.06
P15	0.07	0.29	1.11	0.64	4.90	1.22	28.06	4.38	4.69
P16	0.06	0.39	1.53	0.43	5.44	1.70	22.25	4.80	4.17
P17	0.11	0.27	0.86	0.42	4.40	1.73	22.47	3.85	3.55
P18	0.17	0.67	6.19	1.15	4.26	1.46	31.79	3.66	3.27
P19	0.08	0.48	4.75	1.05	5.55	1.53	26.98	4.95	4.21
P20	0.17	0.37	1.28	0.60	4.62	2.04	25.36	3.83	4.14
P21	0.06	0.41	1.51	0.43	5.44	1.70	22.25	4.80	4.17
P22	0.10	0.47	5.31	1.08	4.57	2.05	26.19	3.84	4.13
P23	0.19	0.73	1.90	0.44	4.26	1.46	31.79	3.66	3.27
P24	0.24	0.94	2.71	0.91	4.36	1.65	26.14	3.42	4.04
P25	0.21	1.04	5.81	1.12	4.46	1.92	27.02	3.57	3.72
P26	0.27	0.91	17.52	1.58	4.02	2.53	21.30	3.22	4.07
P27	0.28	1.11	12.57	1.21	4.03	2.23	28.95	3.28	3.65
P28	0.27	0.96	8.75	2.27	3.89	2.09	19.85	3.07	4.46
P29	0.30	1.18	5.39	1.60	4.41	1.59	21.58	3.94	5.16
P30	0.30	1.37	3.86	1.07	4.89	2.21	17.12	4.32	4.59
P31	0.32	1.28	4.04	1.04	3.96	2.25	17.28	3.47	3.90
P32	0.33	1.18	4.80	1.10	3.83	1.90	24.45	3.30	3.59
P33	0.30	1.19	6.10	1.27	4.99	1.99	20.75	4.45	4.63
P34	0.30	1.34	6.14	1.48	4.14	2.62	19.25	3.48	4.56
Grade	1	1–2	2–3	1–2	3	2	4	3	3

agricultural and other anthropogenic inputs. Only, Cd shows a high degree of contamination.

Table 7 presents the geo-accumulation index for the quantification of heavy metal accumulation in the study area. The I-geo grade for the study area varies from metal to metal and location to location (across metals and locations). Fe remains in grade 0 (unpolluted) in all locations suggesting that the study area soils are in background value with respect to this metal. The I-geo for Mn and Zn attain grade 0 in few locations (unpolluted), while, attain in grade 1 in other soils which indicates that these soils were slightly polluted by Mn and Zn. I-geo index showed that most of (Ni, Co, Cr) heavy metals are in grade 3 (Table 7). Pb and Cu are in grade 2, however, Cd is in grade 6. This

suggests that the soils of Bahr El Baqar are having background concentrations and these elements are practically changed by anthropogenic influences, while the concentration of Cd, Ni Co, and Cr exceeded the average standard value. These dangerous metals may be derived from industrial waste and gasoline additives used, in the factories and cars (Mwamburi 2003). These elements may also be derived through corrosion of the numerous abandoned launches along the drain and agricultural activities.

Combined index

Table 8 shows different combined indices for heavy metal accumulation in the study area. DC values characterize a

Table 7 The geo-accumulation index for the quantification of heavy metal accumulation in the study area

Sampling point	Fe	Mn	Cu	Zn	Ni	Pb	Cd	Co	Cr
P1	−1.90	0.43	1.66	0.45	2.70	1.27	5.27	2.35	2.61
P2	−2.01	0.52	2.18	−0.06	2.88	0.87	5.40	2.72	2.81
P3	−1.71	0.54	2.14	−0.64	3.03	1.35	5.06	2.85	2.65
P4	−2.31	0.11	1.92	−0.67	2.72	1.37	5.07	2.53	2.41
P5	−3.55	0.19	3.01	1.27	2.73	1.25	5.02	2.56	2.95
P6	−1.32	0.42	1.62	0.65	3.06	1.20	5.34	2.89	2.66
P7	−1.29	0.73	1.97	−0.16	2.79	1.62	5.25	2.52	2.63
P8	−4.51	−1.81	0.89	−0.64	3.03	1.35	5.06	2.85	2.65
P9	−4.33	−0.86	−0.47	0.70	2.78	1.62	5.30	2.52	2.63
P10	−3.74	0.46	1.48	−0.59	2.68	1.13	5.58	2.46	2.29
P11	−3.57	0.16	1.72	0.46	2.71	1.30	5.29	2.36	2.60
P12	−2.21	0.67	2.04	−0.07	2.74	1.54	5.38	2.42	2.47
P13	−1.70	0.41	3.20	1.15	2.63	1.97	4.85	2.38	2.77
P14	−2.16	0.30	1.70	0.45	2.70	1.27	5.27	2.35	2.61
P15	−3.32	−1.22	0.73	−0.06	2.88	0.87	5.40	2.72	2.81
P16	−3.42	−0.76	1.20	−0.64	3.03	1.35	5.06	2.85	2.65
P17	−2.61	−1.32	0.36	−0.67	2.72	1.37	5.07	2.53	2.41
P18	−1.98	0.01	3.21	0.78	2.68	1.13	5.58	2.46	2.29
P19	−3.00	−0.46	2.83	0.65	3.06	1.20	5.34	2.89	2.66
P20	−1.97	−0.86	0.94	−0.16	2.79	1.62	5.25	2.52	2.63
P21	−3.55	−0.71	1.18	−0.64	3.03	1.35	5.06	2.85	2.65
P22	−2.80	−0.52	2.99	0.70	2.78	1.62	5.30	2.52	2.63
P23	−1.82	0.13	1.51	−0.59	2.68	1.13	5.58	2.46	2.29
P24	−1.48	0.49	2.02	0.46	2.71	1.30	5.29	2.36	2.60
P25	−1.70	0.64	3.12	0.74	2.74	1.53	5.34	2.42	2.48
P26	−1.31	0.45	4.72	1.25	2.59	1.92	5.00	2.27	2.61
P27	−1.25	0.73	4.24	0.86	2.59	1.74	5.44	2.30	2.45
P28	−1.32	0.53	3.71	1.77	2.54	1.65	4.90	2.20	2.74
P29	−1.15	0.82	3.01	1.27	2.73	1.25	5.02	2.56	2.95
P30	−1.17	1.04	2.53	0.68	2.88	1.73	4.68	2.69	2.78
P31	−1.07	0.94	2.60	0.65	2.57	1.75	4.70	2.38	2.55
P32	−1.02	0.82	2.85	0.73	2.52	1.51	5.20	2.31	2.43
P33	−1.17	0.84	3.19	0.93	2.90	1.57	4.96	2.74	2.80
P34	−1.17	1.00	3.20	1.15	2.63	1.97	4.85	2.38	2.77
Grade	0–1	1	2	1	3	2	6	3	3

very high pollution for all of the Bahr El Baqar soils, reflecting the changes in soil occupation and the intensity of economic activities. The soils no. 26 and 18 is the most contaminated soils which shows the highest DC value of all studied areas (55.41 and 52.61) which classified as very high. The soils no. 31 and 17 show lowest DC values (37.54 and 37.65). Pollution severity and its variation along the sites was determined with the use of pollution load index. This index is a quick tool in order to compare the pollution status of different places. The values of the Pollution Load Indexes (Table 7) were found to be generally polluted (>1) in all the studied soils. The difference in indices results due to the difference in sensitivity of these indices towards the soil pollutants (Praveena et al. 2007).

These confirmed that Bahr El Baqar drain is facing probable environmental pollution, especially with dangerous heavy metals (Pb, Cd, Co, Cr and Ni) which result from increased rate of non-treated industrial waste which are discharged to Bahr El Baqar drain. If this combined index (MPI) is above 1, the concentrations of trace metals would be considered elevated and ecosystem could be regarded as "polluted".

The potential ecological risk index

Table 9 summarizes the results for PERI calculation at the studied areas. CF increment generates an equal increase of DC, since DC = Σ CF, but PERI increment will depend on

Table 8 Different combined indices for heavy metal's accumulation in the study area

Sampling point	DC	MPI	PLI	Sampling point	DC	MPI	PLI
P1	43.30	1.64	2.09	P18	52.61	1.72	2.31
P2	48.04	1.68	2.17	P19	49.57	1.70	2.14
P3	42.90	1.63	2.16	P20	42.41	1.63	1.78
P4	39.78	1.60	1.84	P21	40.76	1.61	1.58
P5	44.49	1.65	2.19	P22	47.73	1.68	2.15
P6	47.46	1.68	2.38	P23	47.70	1.68	1.86
P7	44.59	1.65	2.30	P24	44.40	1.65	2.24
P8	40.25	1.60	1.32	P25	48.88	1.69	2.53
P9	42.74	1.63	1.43	P26	55.41	1.74	2.99
P10	47.71	1.68	1.65	P27	57.31	1.76	2.90
P11	43.52	1.64	1.82	P28	45.61	1.66	2.82
P12	45.95	1.66	2.11	P29	45.15	1.65	2.76
P13	42.76	1.63	2.60	P30	39.73	1.60	2.64
P14	43.25	1.64	2.04	P31	37.54	1.57	2.48
P15	45.35	1.66	1.53	P32	44.48	1.65	2.54
P16	40.78	1.61	1.59	P33	45.66	1.66	2.83
P17	37.65	1.58	1.43	P34	43.30	1.64	2.84
Grade	Very high	Polluted	Polluted	Grade	Very high	Polluted	Polluted

Table 9 Potential ecological risk (PER) values and the results for PERI calculation at the studied areas

Sampling point	Cu	Zn	Pb	Cd	Cr	PERI	Sampling point	Cu	Zn	Pb	Cd	Cr	PERI
P1	10.88	0.93	8.29	796.96	8.36	825.42	P18	31.33	1.16	7.39	965.56	6.61	1012.06
P2	10.75	0.46	4.34	597.69	6.66	619.89	P19	33.56	1.48	10.79	1143.43	11.89	1201.15
P3	17.09	0.50	9.87	774.98	9.69	812.13	P20	4.52	0.42	7.23	538.37	5.86	556.40
P4	11.98	0.40	8.22	641.64	6.75	668.99	P21	5.39	0.31	6.08	477.07	5.97	494.81
P5	29.02	1.73	8.55	697.67	11.11	748.09	P22	17.81	0.73	6.86	526.79	5.53	557.72
P6	8.28	0.85	6.18	655.57	6.82	677.70	P23	6.68	0.31	5.13	669.89	4.59	686.60
P7	11.58	0.53	9.05	673.78	7.33	702.28	P24	10.17	0.69	6.18	588.57	6.07	611.68
P8	4.80	0.33	6.60	518.01	6.48	536.22	P25	25.31	0.97	8.38	706.04	6.48	747.18
P9	3.55	1.59	15.04	1155.78	12.14	1188.12	P26	60.13	1.09	8.67	438.68	5.59	514.16
P10	6.74	0.32	5.27	688.96	4.72	706.01	P27	33.71	0.65	5.97	465.65	3.92	509.89
P11	8.35	0.69	6.25	595.81	6.14	617.25	P28	24.43	1.27	5.85	332.69	4.99	369.22
P12	10.27	0.48	7.28	622.50	5.55	646.08	P29	13.62	0.81	4.01	327.31	5.21	350.96
P13	21.88	1.05	9.33	411.27	6.50	450.03	P30	14.12	0.78	8.09	375.60	6.72	405.31
P14	8.67	0.73	6.44	618.85	6.49	641.17	P31	17.63	0.91	9.80	452.72	6.81	487.88
P15	4.08	0.47	4.51	621.34	6.92	637.32	P32	16.08	0.74	6.36	491.61	4.82	519.61
P16	6.85	0.38	7.61	597.07	7.47	619.39	P33	21.68	0.90	7.06	442.86	6.59	479.09
P17	4.15	0.40	8.36	652.68	6.87	672.47	P34	19.16	0.92	8.17	360.13	5.69	394.07

which metal has this higher CF because it is specific for each one which were classified in decreasing order of toxicity (Cd = 30 > Cu = Pb = 5 > Cr > Zn = 1).

As a predict model, the PERI is dependent on calibration with bio-indicators in order to evaluate its effectiveness for potential risk of soil heavy metals. The PERI application in Bahr El Baqar ecosystems was successful, demonstrating that the environmental variables used in the algorithm proposed by (Hakanson 1980) are the main integrator parameters of biogeochemical processes in this ecosystem. In addition, the relationship among these variables shows a logical synthesis of biogeochemical processes that influence metal behavior in the soils. In this study, spatial distribution of Cd in all study areas was investigated. Cd was chosen because it represents up to 90 % of PERI (Table 9) values for almost all study areas, being representative of metal bioavailability and their risks in these areas.

Table 10 Fitted parameters of the variogram models for heavy metals

Parameters	Models	Prediction errors				
		Mean	Root mean square	Average standard error	Mean standardized	Root mean square standardized
DC	K-Bessel	−0.1088	3.4394	3.9591	0.0051	0.9018
MPI	Exponential	−0.0003	0.0363	0.0388	0.0034	0.9625
PLI	Exponential	−0.0064	0.3253	0.3617	−0.0032	0.9503
Cd (by mean)	Pentaspherical	−0.0875	2.1092	2.2170	−0.0057	0.9914
Cd (by CF)	Rational quadratic	−0.1598	3.5015	3.74614	−0.0076	0.9535
Cd (by I-geo)	Stable	−0.0078	0.2070	0.2158	−0.0077	0.9727
PERI	Pentaspherical	1.3035	185.3357	191.9156	−0.0037	0.9862

Spatial distribution of soil heavy metals

Table 10 and Fig. 3 list the cross validation and fitted parameters results in examining the validity of the different models and parameters of semivariograms (e.g., Cd parameters, single and combined index). Cross-validation was used for comparing the interpolation methods. Three indices were calculated from the measured and interpolated values at each validation sample site. The mean error (ME), the mean absolute error (MAE) and the root mean square error (RMSE) are determined from the measured values. The ME is a measure of the bias of the interpolation, which should be close to zero for unbiased methods, and the MAE as well as RMSE are accuracy measures of the interpolation, which should be as small as possible for accurate interpolation.

For the DC index the best fit is the K-Bessel model (SME 0.0051) and Exponential model for MPI and PLI index with a 0.0034 and −0.0032 SME respectively which is closest to zero. The RMSS values for MPI and PLI are 0.9625 and 0.9603, respectively, which are closest to 1. The best fit for the mean cadmium parameter is the Pentaspherical model (SME −0.0057) and Rational Quadratic model for cadmium (by CF) with a −0.0076 SME which is closest to zero. The best fit for the PERI index is the Pentaspherical model (SME −0.0037). When the average estimated prediction standard errors are close to the root-mean-square prediction errors from cross-validation, you can be confident that the prediction standard errors are appropriate (Johnston et al. 2001).

Table 10 lists cross validation results in examine the validity of the fitting models and parameters of semivariograms for heavy metals. Table 10 shows the most suitable models and their prediction error values for each parameter. Table 10 also shows that for the different parameters, different models may give better results. For heavy metals, RMSS range from 0.9018 to 0.9862. Figures 4 and 5 shows the spatial distribution maps of

different indices (e.g., DC, PLI, MPI, PERI….) in the study area based on these interpolations.

From the point of integrated assessments view, the ecological risk of heavy metals in the surface soils for the study area indicating a high contamination risk which was dominated by Cd (Fig. 5).

Overall discussion

Recall, the principal objective of this study was to assess soil contamination in Bahr El Baqar agricultural area. The discharge of industrial, agricultural and municipal wastewaters in Bahr El Baqar drain has contaminated the environment surrounding Bahr El Baqar areas. The field-work was conducted during the dry season in order to obtain maximal heavy metal concentration from the soil. (Yahaya et al. 2009) confirmed that the concentration of heavy metal in soil is higher in the dry season than in a rainy season because heavier metals are lost in the soil due to run-off and infiltration in a rainy season. The accumulation of heavy metals in these soils is a serious concern due to their persistence and toxicity. Thirty-four samples were collected at 0-20 cm and evaluated for heavy metals using geoaccumulation index (I-geo), enrichment factor (EF) and contamination factor (CF), pollution load index (PLI), and metal pollution index (MPI), etc.

The concentration of cadmium ranges from 10.27 to 19.07 mg/kg with a mean concentration of 14.69 mg/kg. The calculated geo-accumulation index (I-geo) for cadmium indicates that the soils are extremely contaminated and very high contamination degree with the CF index. Cd is regarded as one of the most toxic trace elements in the environment. Cd is higher in the study area because of the uses of phosphate fertilizers, irrigation by untreated wastewater of Bahr El Baqar darin. Therefore, the water from this drain is not suitable for agricultural purposes. Manganese ranged from 13.73 to 98.73 mg/kg. The mean was 58.98 mg/kg. The calculated I-geo value gave a value
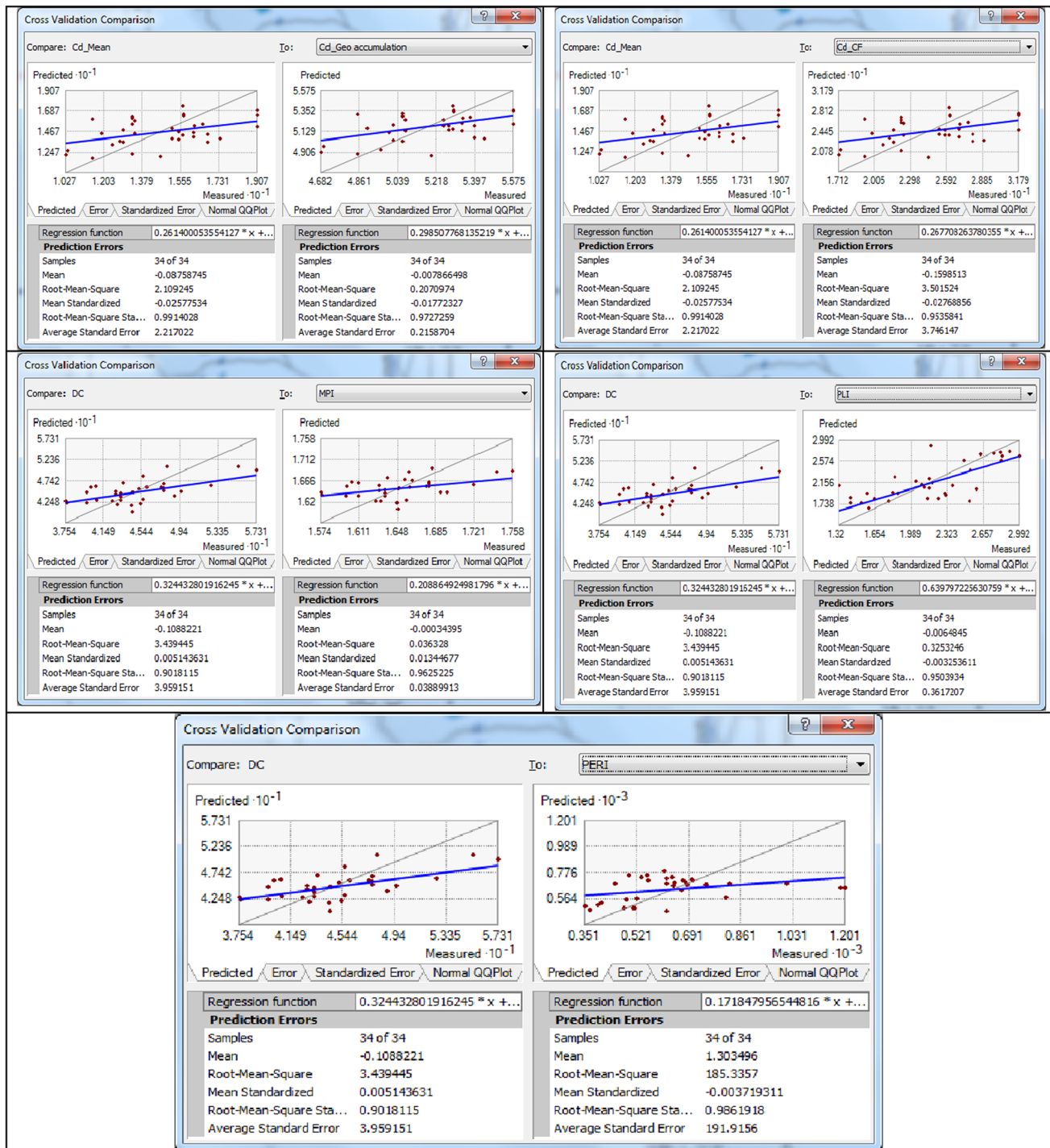
Fig. 3 The cross validation comparison of the soil heavy metals map by different methods

that indicates uncontaminated to moderate and low contamination degree with the CF index. Manganese can be adsorbed onto soil depending on organic content, pH, grain-size and cation exchange capacity of the soil and this can be exemplified by the strong positive correlation with organic matter (<0.01 level). The concentration of copper varied from 7.73 to 280.30 mg/kg with an average value of 65.70 mg/kg. A moderately positive, high correlation with lead and Zinc was established (<0.01 level). Although zinc remains adsorbed to soil, leaching has been reported at waste disposal sites. The lower concentrations of the Zn might be due to the continuous removal of heavy metals by
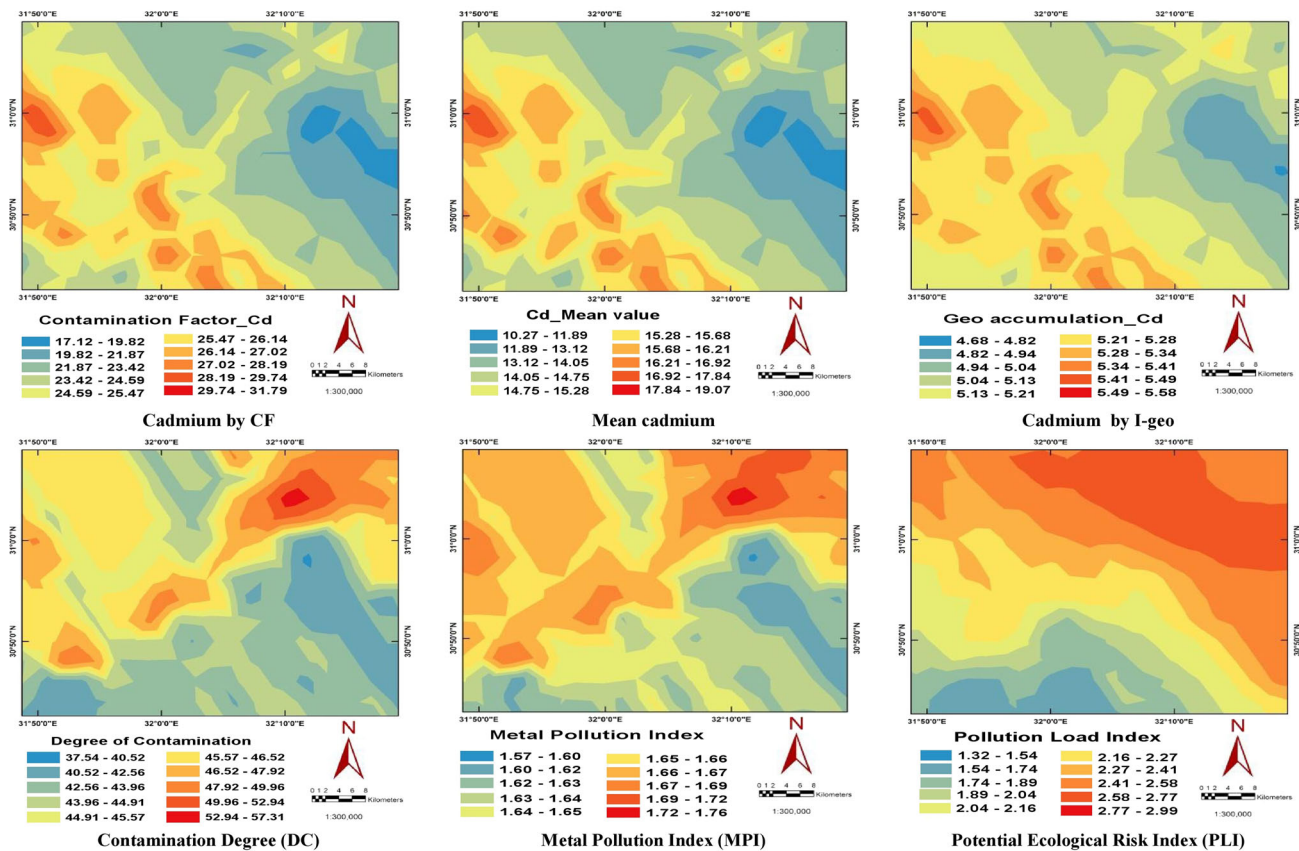
Fig. 4 Spatial distribution maps of different models

the crops grown and due to leaching of heavy metals into the deeper layer of the soil and in to the ground water. Chromium concentration ranges from 84.92 to 134.06 mg/kg with a mean value of 106.96 mg/kg. Chromium may be lower in some sites due to the continuous removal of heavy metals by the crops and due to leaching of heavy metals into the deeper layer of the soil and to the ground water. No correlation was found with other metals and its concentration falls within the contaminated. Nickel measured concentrations (61.32–88.73 with mean 73.22) are above the average reference abundance in an uncontaminated soil. A moderate positive correlation with Zn was noted at <0.05 level. The results show that lead concentration ranged from 24.42 to 52.40 mg/kg with a mean concentration of 36.64 mg/kg. Though there was an observed strong correlation with Cu (<0.01 level), its concentration is within the level of uncontaminated soil. Zn concentration varied between 39.67 mg/kg and 215.58 mg/kg with an average concentration of 90.56 mg/kg. These values are found to be low the average abundance for an uncontaminated soil. The I-geo concentration lies below the range of uncontaminated soil. Zinc had very strong positive correlation with Cu and Pb (<0.01 level) and moderate positive correlation with Ni (<0.05 level). The irrigation by Bahr El

Baqar wastewater and uses of agricultural fertilizers led to the increasing the Ni concentrations. Cobalt concentration ranged 70.51–113.82 mg/kg with a mean value of 89.72 mg/kg. The measured concentrations of Co are in range of contaminated soil. The irrigation of agricultural lands with untreated water led to the accumulation of Co in the soils. Among significant variables that control the distribution and enrichment of heavy metals in the soils are soil pH, grain size of the soil, amount of organic matter in the soil and the cation exchange capacity (Huang and Lin 2003; Lin et al. 2002). The soil pH is generally high (7.58–8.82) while $CaCO_3$ (0.61–29.2) characterize the top soil and these condition enhances the precipitation and bio-accumulation of heavy metals in soil. Heavy metals have a strong affinity for organic content, clay and silt fraction because of their high cation exchange capacity (Bodur and Ergin 1994; Zonta et al. 1994). The topsoil comprises organic content (0.22–3.55), clay and silt fraction.

Four principal components (Eigenvalues >1) emerged accounting for 78.11 % of the cumulative variance from the principal component analysis. The first principal component (PC-1) loading with 47.24 % variance showed higher loading for Cd, Cu, Co, Fe and organic matter. The second principal component (PC-2) has loading 13.65 % of

Fig. 5 Prediction maps of the potential ecological risk index (PERI)
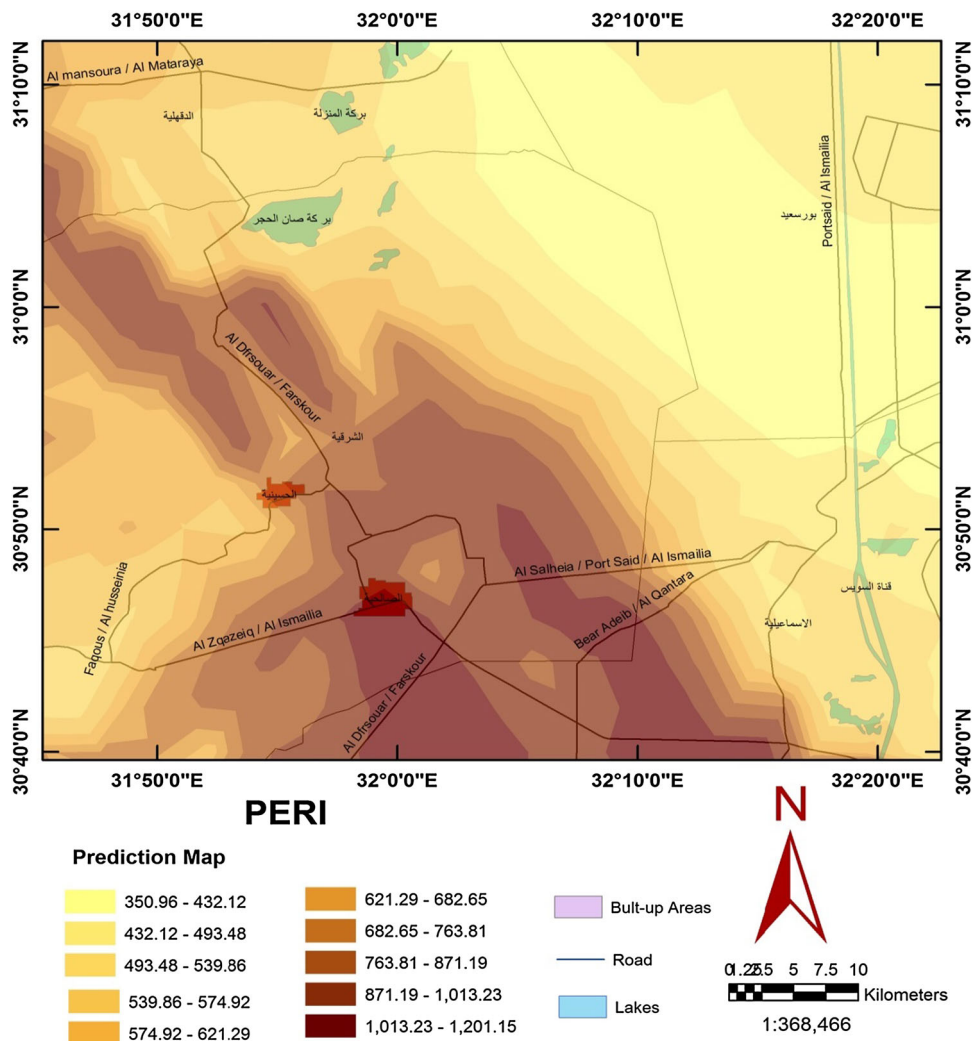
the total variance, had high loading. The pH of the soil could have contributed to Pb and Zn retention in the soil, resulting in low mobility of the metals (Amadi et al. 2012; Yoshida et al. 2002). The third principal component (PC-3) explains 10.72 % of the total variance and consists of silt, clay, $CaCO_3$, CEC, OM and pH. The fourth principal component (PC-4) has a moderate loading for Mn, and Zn, which accounts for 6.49 % of the total variance. Industrial activities domiciled in the area may be responsible for the presence of Mn and Zn. The physico-chemical properties of clay could have encouraged their availability in the soil. By applying the Pearson rank order correlations, Fe is well correlated with Mn and moderately correlated with Cu and Zn. There is a good correlation between Ni and Co. There are significant correlations between Cu and each of Zn and Pb. Ni, Fe, Mn, Cu, Zn and Co are positively correlated with OM, pH, clay, $CaCO_3$, and CEC and of course negatively correlated with sand. These results revealed that these heavy metals in the soils have the same source of contamination, which is Bahr El Baqar drain.

Conclusions

This work provides a comprehensive analysis of heavy metal assessment in Bahr El Baqar surface soils. Different indices have been employed for the evaluation of heavy metal contamination status. The impact of anthropogenic heavy metal pollution in the sampling locations was evaluated using Enrichment Factor (EF), Contamination Factor (CF), Contamination degree, Pollution load Index, Metal Pollution Index (MPI), and Geoaccumulation Index (I-geo) at 34 sampling locations of Bahr El Baqar region. The results showed that average CF values for heavy metals have an order Cd > Cr > Co > Ni > Cu > Pb > Zn > Mn > Fe, suggesting that soil samples was extremely highly enriched with Cd, while Pb exhibit significant enrichment. The results of EF of all sampling sites were found to be less than 2 indicates the study area falls in the category of deficiency to low enrichment. The results of Contamination Factor, and Contamination degree, show that the study area falls under moderate to very high

contamination degree. Calculation of the pollution load Index, Metal Pollution Index, Metal Contamination Index and Geo accumulation Index indicates the study area is regarded as polluted. The present study suggests that these indices are useful tools for identification of anthropogenic source of soil contamination. Multivariate analysis and correlation matrix were used in this study. A significant positive correlation is observed among Fe and Mn, and moderately correlated with Cu and Zn. There is a good correlation between Ni and Co indicating that these metals were derived from similar sources. Ni, Fe, Mn, Cu, Zn and Co are positively correlated with OM, pH, clay, $CaCO_3$, and CEC and of course negatively correlated with sand. From the point of integrated assessments of multiple heavy metal pollution, the ecological risk of heavy metals in the surface soils in the study area indicating a high risk, which was dominated by Cd. This study shows that although there were variations in the results of the different indices, the combination indices gave us a comprehensive understanding of heavy metal risks in the surface soils of the Bahr El Baqar region.

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