

Ecological and Health Risk Assessment of Trace Metal Pollution in Vegetable Field Soils from the Eastern Nile Delta, Egypt

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Abstract

The accumulation of trace metals in vegetable field soils is of expanding worry because of the potential health hazards and its detrimental effects on soil ecosystems. To investigate the state of trace metal pollution in vegetable field soils, 60 surface soil samples were collected from vegetable fields across the Eastern Nile Delta region, Egypt. The results explained that the concentrations of Cu, Mn and Ni were lesser than their corresponding background values, while the concentrations of Cd, Co, Pb and Zn were exceed their background values. The pollution indices showed that the study soil experienced low to moderate contamination, and the Cd and Cr contamination was serious. The hazard index values of nine trace metals signified that there no adverse non-carcinogenic risk for adults and children. The carcinogenic risk of Cd, Co, Ni and Pb for both age groups was within the acceptable limits, while Cr had critical carcinogenic hazard to children. Overall, the quality of studied soils is relative safety, although some samples impose serious pollution problems by Cd and Cr. Thus, properly monitor trace metals and soil management action should be applied to reduce further soil pollution in vegetable fields in the Eastern Nile Delta.

Introduction

Over the last few decades, the agricultural soil pollution by trace metals has received considerable attention because of their toxicity at low content, bioaccumulation and persistence^{1,2}. The accumulation of trace metals in agricultural soil can threaten the environmental ecosystem safety such as decline the soil and groundwater quality³. Moreover, it can threaten food security by adverse effects on the crop quality and the health of animals. These might cause potential risks to the human health through the food supply chain and groundwater consumption^{4,5}. The trace metals of agricultural soil can mainly enter the human body through three exposure routes: ingestion, inhalation and skin contact⁶. The migration and accumulation of trace metals in agricultural soils are associated with both the chemical and physical properties of soils⁷. The agricultural soil is classified as polluted when the trace metals concentration in its bulk horizons is higher than the baseline values taken as upper limits for no contaminated soils⁸.

Generally, the trace metals in agricultural soils come from natural processes and/or anthropogenic activities². The natural origin of trace metals is related to soil parent materials⁹. The contribution of anthropogenic activities to the trace metals in rural soils was a lot more than the natural source¹². Agricultural activities especially the utilization of organic and inorganic fertilizer, the use of large quantities of pesticides, and the use of low quality water are the major sources of trace metals caused by anthropogenic activities¹⁰. The large input of agrochemical leads to soil degradation and pollution especially in vegetable fields¹¹. Soil pollution pose an important threat to food security by both decreasing crop yields because of the toxicity of pollutants, and by causing unsafe crop yields for animals and humans. When contaminations exceed natural levels in the soil, not only soil degradation is occurring, but crop yields and human health can also be influenced¹². Thus, it is essential to estimate the potential health risk associated with trace metal pollution in agricultural soils. Moreover, as well as imperiling human health and the environment, soil contamination can likewise lead to serious economic losses¹³.

The Nile Delta of Egypt covers a large area of agricultural land and represents a vital economic sector. The continuous urbanization and industrialization of the Nile Delta and surroundings have prompted a raise in the pollution of water resources and soils, causing a probable health hazard¹⁴. The quick increase in soil contamination in Egypt has turned into a real danger to the people's health and economy. Egyptian farmers had shifted use of their land for crop production in several seasons per year. Thus, they apply excessive amounts of mineral fertilizers and pesticides without any guidelines to increase crop productivity and reduce crop losses, especially in the Nile Delta¹⁵. Moreover, the domestic wastewater is reused for agricultural irrigation in rural areas in this region¹⁶. These practices have been causing the aggregation of trace metals in soils and the degradation of agrarian soils^{17,18,19,20}.

Several studies have been carried out over the past few on trace metal pollution of agrarian soils and ecological risk in soils of different regions in Egypt. The Nile water receives a lot of pollutants from wastewater discharging through agricultural drains and industrial sources. Thus, the trace metals pollution transferred from the water to the soil and accumulated with time^{19,21-24}. Sometimes, farmers depend on drainage water for irrigating their soils in such an area^{20,25}. These indicate that agricultural soil pollution might be influenced by human activities and require more attention in the future.

Pollution indices have been widely and efficiently applied to provide a comprehensive description of the status of trace metal contamination in soil. Shokr et al.²⁶ found that the pollution load index was > 1 in most soil samples of the middle part of the Nile Delta, showing considerable to high polluted classes with trace metals. Omran¹⁷ found that pollution indices revealed that the soils of Bahr El Baqar area fall under moderate to very high load of trace metals. Khalifa and Gad²³ found the highest degrees of pollution and potential ecological risk in the soils of Quessna district in the Southwestern Nile Delta. Salman et al.⁴ found that the pollution indices revealed that the soil samples from Orabi farms, El Obour city were fluctuated from considerable pollution as per the potential ecological risk index values to high polluted pattern as per the pollution load index and pollution degree results. Abou El-Anwar, et al.¹⁹ found hat the pollution indices explained that the agricultural soils at Aswan and Luxor can be considered generally moderately polluted with heavy metals, but they have a very high ecological risk with Cd. However, studies on heavy metal pollution of agricultural soils and ecological risk, particularly in the Eastern Nile Delta ecosystems remain limited.

The pollution characteristics of trace metals in agrarian soil might vary with various types of crops, particularly when their production continuous for many years⁹. The agrochemicals are applied at higher rates in vegetable crops than in most others. Excessive amassing of trace metals in vegetable field soils through the long-term application of agrochemicals and by other sources may result in soil pollution²⁷. Trace metals accumulate at high level in the vegetable edible parts when contrasted with fruit or grain crops. The consumption of vegetables cultivated in soils that are contaminated with trace metals can cause clinical problems to human health¹⁶.

In Egypt, the studies on trace metal pollution of soil in vegetable fields are very limited on large or moderate regional scales. Moreover, the studies on associated human health risks are not common. Further studies for human health risk evaluation are needed to explore potential risks for humans from the pollution of trace metals in agrarian soils of the Eastern Nile Delta area²³. Thus, monitoring the status of trace metal pollution in the soil of vegetable areas is

necessary for estimating the potential health risk and adequate management of these pollutants as well as promoting food safety. Thus, the three main objectives of this study were: (1) to determine the concentrations, sources and spatial distributions of trace metals in the vegetable field soils of the Eastern Nile Delta area, (2) to assess the trace metal pollution of vegetable field soils and ecological risk, and (3) to evaluate the potential health risk of the trace metals based on various exposure routes.

Results And Discussion

Descriptive statistical analyses.

Trace metals.

The descriptive statistics of the content of trace metals in the vegetable field soils from the Eastern Nile Delta are given in Table 1. The descriptive statistics results show that the mean concentrations of Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn were 2.36, 16.12, 151.56, 9.85, 20972.74, 479.63, 27.25, 28.31 and 100.01, with a median concentration of 2.78, 19.98, 178.90, 10.48, 25634.40, 576.30, 29.68, 31.30 and 116.91 mg kg⁻¹, respectively. The SD values were high because the concentrations of trace metal had quite high heterogeneous distribution in the studied area.

Table 1

Descriptive statistics of trace metal concentrations (mg kg⁻¹) and soil properties (clay %, organic matter % (OM), pH, and electrical conductivity (EC) dS m⁻¹) in the soils of the study area. K-S: Kolmogorov-Smirnov test, CV: coefficient of variation, BG: Background as reported by Kabata-Pendias (2011), Avg: average, s: standard deviation, cv: coefficient of variation, max: maximum value, min: minimum value, skew: skewness, Q1: lower quartile, middle quartile (median), Q3: upper quartile, MAC: Ranges of Maximum Allowable Concentrations for trace metals in agricultural soils (mg kg⁻¹) (Kabata-Pendias, 2011).

Variables	Minimum	Q1	Median	Mean ± S.D.	Q3	Maximum	CV %	Skewness	Kurtosis	K-S	MAC	Background values
Cd	0.17	2.06	2.78	2.36±1	3.02	4.86	47.26	-0.53	-0.19	0.19	1-5	0.41
Co	1.65	4.32	19.98	16.12±9	22.55	28.43	55.00	-0.66	-1.24	0.24	20-50	11.2
Cr	37.27	58.19	178.90	151.56±70.16	205.24	255.05	46.29	-0.60	-1.22	0.21	50-200	
Cu	2.09	5.86	10.48	9.85±5	13.33	18.04	45.95	-0.08	-1.01	0.11	60-150	38.9
Fe	2162.20	6201.24	25634.40	20973±11553	29016.30	38350.88	55.09	-0.70	-1.13	0.24	-	-
Mn	68.28	135.24	576.30	479.63±267	677.72	935.98	55.67	-0.53	-1.23	0.19	-	480
Ni	12.18	18.74	29.68	27.25±8	33.06	40.04	29.02	-0.33	-1.23	0.15	20-60	29
Pb	0.00	17.11	31.30	28.31±16	40.13	60.90	56.28	-0.06	0.84	0.10	20-300	27
Zn	0.00	51.18	116.91	100.01±41	126.76	170.30	41.15	-0.62	-0.50	0.18	100-300	70
Clay	0.70	8.15	38.40	31.17±19	44.81	61.74	60.52	-0.57	-1.12	0.18	-	-
OM	0.42	0.83	1.14	1.21±0.5	1.65	2.21	40.99	0.23	-1.04	0.10	-	-
pH	7.10	7.80	8.10	8.02±0.4	8.30	8.70	4.69	-0.70	-0.07	0.15	-	-
EC	0.98	1.42	1.85	2.07±0.9	2.42	5.02	42.94	1.49	2.22	0.13	-	-

Omran¹⁷ found that the ranges of Co (70.5-113.8), Cu (7.7-280.3), Ni (61.3-88.7) and Zn (39.7-215.6) concentration (mg kg⁻¹ soil) in the soils of Bahr El Baqar in the Eastern Nile Delta tended to be upper than the ranges found in this study, while the ranges of Cr (84.9-134.1) and Pb (16.4-52.4) tended to be low. The close difference between the lower quartile and the upper quartile points to the distribution and uniform source of trace metal in soil. Among nine trace metals, Cd had a relatively close difference between these quartiles. This may be due to more adding of P-fertilizers that contain a significant amount of Cd, about 12.45 mg kg⁻¹¹⁸. Moreover, cadmium may also be added to soils adjacent to roads²⁸.

Among all trace metals, Fe has the largest mean value (20973 mg kg⁻¹), while Cd has the lowest mean value of 2.36 mg kg⁻¹. These results were consistent with the mean values of Fe and Cd found in agricultural soils in Egypt²². The mean concentrations of Cd, Co, Cr, Cu, Ni, Pb and Zn were below the maximum value of the range of Maximum Allowable Concentrations (MAC) mentioned by Kabata-Pendias²⁹. The mean concentrations of Cu, Mn and Ni were lower than their corresponding background values. However, the mean concentrations of Cd, Co, Pb and Zn were exceeded their background values, which can demonstrate that the soils of the studied area are polluted with these metals by the anthropogenic activities²³. In the same line, higher contents of Cd, Pb, Cu, Zn were observed in vegetable fields in China³⁰. The main contribution of trace metals to agricultural soils is the use of chemical fertilizers, pesticides and organic fertilizers^{5,17,30}. Thus, Cd, Co, Pb and Zn require more attention in the future, and continuous monitoring of the application of agrochemicals in the production of vegetables is recommended to decrease the probable ecological risks caused by these trace metals in soils of the Eastern Nile Delta.

The coefficient of variation (CV) measures the variability degree of the trace metal concentrations in soils. A high value of CV indicates that the contamination of soils by trace metal is produced mainly from human activities. A low CV value indicates that trace metal pollution is due to natural sources^{1,2,31}. The high concentrations of metals coupled with high variation suggest that anthropogenic inputs may be their primary source in the study area. The highest CV values were recorded for Cd (47.26), Co (55.00), Cr (46.29), Cu (45.95), Fe (55.09), Mn (55.53), Pb (56.28) and Zn (41.15). The high CV value confirms that these metals have a wide concentration range and is an indication of the potential impact of human activities on them. Lower values of CV for Ni (29.02) confirm that human activities are less effective on it³¹.

The skewness values of Cu and Pb were near zero and came near a normal distribution, while the other metals had slightly negative skewness, indicating that the center of distribution is shifted to the right. The kurtosis of Pb was greater than zero, indicating the distribution has a towering shape. The kurtosis values of other elements were negative, indicating that the distribution has a flat shape. The results of the Kolmogorov–Smirnov test ($p < 0.05$) confirm that the concentrations of Cu, Ni and Pb are nearly normal distribution, while the concentrations of other metals did not follow a normal distribution.

Soil physicochemical parameters.

The descriptive statistics analyses of soil physicochemical parameters, *i.e.*, clay, OM, pH and EC are given in Table 1 show that the study area had a wide range of contents of clay (0.70–61.74; median: 38.40) and EC (0.98–5.02; median: 1.85 dS m⁻¹). The soils were moderately alkaline with mean pH of 8.02, with low OM (mean 1.21), as typically found in the Mediterranean environments. The CV values were highest for clay (60.52%), OM (40.99%) and EC (42.94%), indicating a non-homogenous distribution. The soil pH had a homogeneous distribution of variables, as indicated by the low value of CV of 4.69%. These results are in line with those found by Aitta et al.²⁰.

The soil clay and pH observations skewed to the right, while the OM and EC observations skewed to the left. The Kurtosis results showed that clay and OM parameters revealed a flat shape, while the distribution of EC values exhibited a towering shape. The pH observations were subject to normal distributions. The K-S test suggested that clay values were not normally distributed, while OM, pH and EC values approached a normal distribution.

Multivariate statistical analysis.

Correlation analysis.

The correlation between different variables is presented in Table 2. The relationship that occurs between trace metals in the soils is usually due to parent material, the influence of pedogenic process, and the effect of human activities³². Very significant positive correlations were found between all trace metals except Cu, which indicated that Cd, Co, Cr, Fe, Mn, Ni, Pb and Zn could be largely derived from the same sources and spreading³². The Cu concentration was positively and significantly correlated with the concentration of Fe, Mn and Ni. Non-significant correlations of Cu with Cd, Co, Pb and Zn may be due to the various processes like external inputs and biological effects. A similar trend was reported in the Southwestern Nile Delta, where the associations between Cr, Cu, Ni, Pb and Zn and scavenger metals (Fe and Mn) had significant high associations²³.

Table 2
The Pearson correlation coefficients between trace metals and soil properties. * $p < 0.05$, ** $p < 0.01$

Variables	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Clay	OM	pH
Cd	1											
Co	0.780**	1										
Cr	0.763**	0.957**	1									
Cu	0.097	0.178	0.202*	1								
Fe	0.804**	0.986**	0.951**	0.216*	1							
Mn	0.735**	0.979**	0.953**	0.201*	0.982**	1						
Ni	0.665**	0.942**	0.905**	0.202*	0.948**	0.952**	1					
Pb	0.576**	0.664**	0.668**	-0.017	0.579**	0.601**	0.496**	1				
Zn	0.589**	0.676**	0.665**	0.161	0.651**	0.614**	0.583**	0.655**	1			
Clay	0.732**	0.895**	0.880**	0.212*	0.905**	0.895**	0.816**	0.634**	0.709**	1		
OM	0.556**	0.774**	0.763**	0.321**	0.777**	0.775**	0.796**	0.504**	0.549**	0.688**	1	
pH	0.288**	0.410**	0.483**	-0.059	0.396**	0.402**	0.356**	0.497**	0.510**	0.531**	0.345**	1
EC	0.371**	0.400**	0.455**	0.017	0.387**	0.416**	0.359**	0.456**	0.240*	0.320**	0.251*	0.365**

The correlation among trace metal concentrations and soil properties is presented in Table 2. Clay % had very significant positive correlations with Cd (0.732), Co (0.895), Cr (0.880), Fe (0.905), Mn (0.895), Ni (0.816), Pb (0.634) and Zn (0.709), and had significant positive correlations with Cu (0.212). The high degree of correlation endorse that, the clay is acting as metal carrier and play a vital role in the distribution pattern of trace metals²³. The positive relationships between all studied trace metals and organic matter percentage were very highly significant. The soil properties especially soil organic matter and clay particles content effectively adsorb trace metals^{23,40}. Soil organic matter is one of the main soil properties with dual effects on trace metals mobility⁵. This

may be because organic matter decomposition produces organic chemicals into soil solution that might act as chelates and raise the bioavailability of trace metals. However, the organic matter of the clay fraction might also decrease the trace metal bioavailability by forming stable complexes with humic substances or through adsorption³³. Both pH and EC correlated positively with Cd, Co, Fe, Mn, Ni, Pb and Zn. Significant correlations were not found between Cu and soil pH and EC. A similar finding has been found by Aitta et al.²⁰.

Factor analysis.

Factor analysis was carried out to identify the sources of pollution. The results of factor analysis using the varimax rotation method are presented in Table 3. Four factors with eigenvalues > 1.0 were selected for the retention data of trace metals in the studied area, which contributed to approximately 87.41% of the total variance. The first factor explains 50.43% of the total variance with negative loadings on all elements. The first factor was characterized by high loadings (≥ 0.72) of Co, Cr, Fe, Mn and Ni, and moderate loading (0.520) of Cd. Several studies found that most of these metals originated from parent material in agricultural soils³². Thus, this factor may represent a natural source. Factor 2 accounting only for about 11.50% of the total variability. It was heavily loaded (≥ 0.99) with Cu with positive loading. The increase Cu level in soil most probably due to the agricultural activities such as fertilizer and fungicide application^{34,35}. Factor 3 explained 13.28% of the total variance. It mainly condensed the information of Pb with negative loadings (- 0.867). The higher content of Pb might have come from anthropogenic activities (2). The fertilizers and traffic emissions that can be the major source of the amount and distribution of Pb in agricultural soils³¹. Factor 4 exhibits 12.20% of the total variance with positive loading (0.852) on Zn. Organic manure and phosphate fertilizers cause a significant expansion in levels of Zn³⁶.

Table 3
Factor loadings rotated matrix for trace metals.

Variables	Factor 1	Factor 2	Factor 3	Factor 4
Cd	-0.520	0.017	-0.227	0.217
Co	-0.876	0.065	-0.287	0.250
Cr	-0.847	0.093	-0.305	0.232
Cu	-0.120	0.991	0.028	0.051
Fe	-0.885	0.130	-0.186	0.233
Mn	-0.915	0.087	-0.241	0.186
Ni	-0.942	0.081	-0.128	0.200
Pb	-0.346	-0.051	-0.867	0.278
Zn	-0.377	0.078	-0.302	0.852
Eigen value	4.54	1.035	1.20	1.10
Variance%	50.43	11.50	13.28	12.20
Cumulative (%)	50.43	61.93	75.21	87.41

The spatial and vertical distribution of trace metals levels are influenced by soil properties, for example, content of clay and OM. Industrial activity, agricultural practices and intensive urbanization are the primary anthropogenic sources of trace metal pollution. Cr, Cu, Ni, Pb and Zn are derived from uncontrolled utilization of phosphate fertilizers, and by atmospheric deposition from industrial activity and urban areas^{2,37}, while Co is enhanced by the organic manure application to the agricultural soil^{51,55}. Cu and Cr are produced from reactions involving Cu SO_4 , which mainly originated from disease prevention^{34,35}. The results of factor analysis also indicate that Fe and Mn came from different potential sources. Agricultural soils can be polluted with Co, Cr, Ni and Zn that might be mainly originated from the application of fertilizers and organic manure^{17,29,51,55}. Moreover, it can be polluted with Cr, Cu, Ni, Pb and Zn that result from industrial activity and atmospheric deposition^{24,55}. Atmospheric deposition from industrial and urban areas represented 80.36% of Zn, 76.55% of Pb, 67.48% of Cu, 62.23% of Cr and 37.79% of Ni entering agrarian soil from anthropogenic activities³⁷. It appears to be that anthropogenic and lithologic practices are the chief sources of Cd accumulation in soil. Cadmium is a metal marker of agricultural production activity, and it can be come from atmospheric deposition^{30,34}.

Cluster analysis.

The results of the cluster analysis show that nine trace metals in the soils of the Eastern Nile Delta were divided into five categories (Fig. 1). The first cluster contained Cu. The second cluster contained Zn. Cluster third contained Pb. The fourth cluster contained Cd. The fifth cluster contained Mn, Fe, Co and B. The clustering results were in agreement with the factor analysis results.

Spatial distribution of trace metals.

The spatial distribution maps of trace metal contents in the vegetable field soils of the Eastern Nile Delta are presented in Fig. 2. Generally, the concentrations of all studied trace metals were the highest in the northwest part of the region. The possible reason for this is that some vegetable fields are close to industrial regions and intensive traffic activity, and irrigated with wastewater, which leads to the higher soil trace metal contents. Additionally, very high values of Co, Cr, Cu, Fe, Mn and Ni were also recorded in the central part. The sites with the lowest concentration of most trace metals are located in the southeast part of the

region, which suggests that vegetable soils in this part might not have been affected by industrial activities. The distribution pattern of the Cd and Cu are mainly similarly distributed over the study region. This suggests the primary role of agricultural activities such as fertilizer and pesticide application in the vegetable fields as the main pollutant sources.

Pollution assessment of trace metals.

The EF values for the studied metals are presented in Fig. 3. The EF values indicated that Co, Cu, Fe, Mn and Ni < 2, showing no enrichment. The metals of Cr, Pb and Zn indicated moderate enrichment with EF mean of 4.856 4.773 and 3.874 respectively. Khalifa and Gad²³ and Abou El-Anwar²¹ found similar results. The maximum EF values was 22.687 for Cd and consequently signifying very high enrichment. The EF values < 2 point to the trace metal is completely come from a geological origin, but the EF values > 2 indicate that the trace metal possibly derives from anthropogenic activities³⁸.

The values of geo-accumulation index (*Igeo*) for the studied metals are illustrated in Fig. 3. Generally, the positive *Igeo* values indicate that the metal contamination is related to anthropogenic activities³⁹. The mean values of *Igeo* increased in the order of Fe (0.0002) < Mn (0.007) < Zn (0.0450) < Cu and Ni (0.046) < Co (0.123) < Pb (0.145) < Cd (2.047) < Cr (3.114). The range of *Igeo* values for individual metal is as follows: Cr (0.039 - 0.059), Cd (5.705 - 5.068), Co (0 - 0.169), Cu (0.016 - 0.062), Fe (0.00016 - 0.00022), Mn (0.005- 0.008), Ni (0.035- 0.052), Pb (-0.012 - 0.198) and Zn (0 - 0.052). Based on the mean values of *Igeo*, Cr had trace contamination, and Cd had moderately to heavily contaminated. However, the other metals showed uncontaminated to moderately pollution load. This result is consistent with one previous review on agricultural soils, which shown that the *Igeo* values of Cr and Cd were above 1^{6,23,34}.

The computed results of the contamination factor (*CF*) for studied trace metals are presented in Fig. 3. The mean *CF* values of trace metals decreased in the following order: Cd (7.851) > Cr (1.684) > Pb (1.416) > Zn (1.053) > Co (0.848) > Mn (0.564) > Fe (0.444) > Ni (0.401) > Cu (0.219). The mean *CF* value for Cd indicated a very high contamination level (*CF* > 6), while the mean *CF* values for Cr, Pb and Zn showed a moderate contamination level (1 < *CF* < 3), and the mean *CF* values for Co, Mn, Fe, Ni and Cu pointed to a low contamination level (*CF* < 1). *CF* show minor similarity with Omran's¹⁷ study in the soils of Bahr El Baqar in the Eastern Nile Delta, Shokr et al.²⁶ in the soils of the middle Nile Delta, and Abou El-Anwar²¹ in the soils of the Upper of Egypt.

Health risk assessment.

The values of *HQ*, *HI*, and *CR* related to nine metals for both adults and children are summarized in Table 4. The *HQ* values of different metals through three pathways reduced in the order of ingestion > dermal absorption > inhalation. This result implies that ingestion of soil particles is the major route for trace metals that were adverse to human health. Previous studies have obtained similar results^{10,35}. The *HI* values of soil trace metals for both adults and children were far lower than the safe level (*HI* ≤ 1) and reduced in the following order: Zn > Cr > Mn > Fe > Cd > Pb > Ni > Co > Cu. These results indicated that the non-carcinogenic threat for children and adults is relatively light across the vegetable field soils of the study area.

Table 4
Health risks of trace metals in vegetable soils from the Eastern Nile Delta.

Metals	<i>HQ</i> _{ing}		<i>HQ</i> _{derm}		<i>HQ</i> _{inh}		<i>HI</i>		<i>CR</i>	
	Adults	Children	Adults	Children	Adults	Children	Adults	Children	Adults	Children
Cd	1.61E-03	3.61E-03	1.29E-03	8.43E-03	3.44E-05	8.30E-05	2.93E-03	1.21E-02	2.17E-09	5.23E-09
Co	5.52E-04	1.24E-03	5.51E-06	3.61E-05	4.12E-04	9.95E-04	9.70E-04	2.27E-03	2.31E-08	5.57E-08
Cr	3.46E-02	7.75E-02	1.38E-02	9.04E-02	7.74E-04	1.87E-03	4.92E-02	1.70E-01	9.30E-07	2.24E-06
Cu	1.69E-04	3.78E-04	4.49E-06	2.94E-05	3.58E-08	8.64E-08	1.73E-04	4.07E-04	-	-
Fe	2.05E-02	4.60E-02	1.64E-04	1.07E-03	3.83E-06	9.24E-06	2.07E-02	4.70E-02	-	-
Mn	7.14E-03	1.60E-02	1.42E-03	9.28E-03	4.90E-03	1.18E-02	1.35E-02	3.71E-02	-	-
Ni	9.33E-04	2.09E-03	2.96E-05	1.94E-04	1.93E-07	4.66E-07	9.63E-04	2.28E-03	3.34E-09	8.07E-09
Pb	5.54E-03	1.24E-02	2.95E-04	1.93E-03	1.17E-06	2.84E-06	5.84E-03	1.43E-02	1.65E-07	3.70E-07
Zn	2.28E-04	5.11E-04	9.11E-06	5.97E-05	4.87E-08	1.18E-07	9.42E-02	2.85E-01	-	-

The *HI* and *CR* values for adults were lower than values for children. Previous studies reported that children experienced higher hazards by trace metal contamination than adults³⁵. The *CR* of Cd, Cr, Ni and Pb were low than the safe value (1×10⁻⁶) and had no risk⁴⁰. The *CR* values for Cr were between 1×10⁻⁶ and 1×10⁻⁴ for children, which indicate that a lower but elevated carcinogenic risk. Therefore, children have much more chances of carcinogenic risk from Cr exposure in the study area than adults. Similar results were found by Song et al.⁴¹, Mo et al.⁷ and Liu et al.³² who reported that Cr posed a significant carcinogenic risk. Moreover, previous studies have revealed that human exposure to low Cr concentrations for long-term can cause poisonous and cancer-causing impacts in people⁴². Also, Zhao et al.³⁵ and Liu et al.³² reported the Cr in soil caused a significant carcinogenic risk to adults and children. Thus, particular attention should be paid to Cr pollution. Risks associated with non-carcinogenic and cancer results from the investigation region were overall lower than those found in the rural areas irrigated with wastewater in the Nile Delta⁴⁰.

Materials And Methods

Study area.

The study area is located between latitudes 30° 35' and 31° 29' N and longitudes 31° 18' and 32° 04' E (Fig4). It covers a major part of the eastern portion of the Nile Delta, including Damietta, Dakahlyia and El-Sharkia Governorates. It has a hot arid summer and low rainy winter, with the total yearly precipitation is around 167 mm/year that falls mainly between October and March. The average annual temperature ranges are 24-35°C in summer and 8-20°C in winter. The soil types are varied from clay to sandy soils, which have high pH values. The Damietta branch of the Nile River is the main source of fresh water for irrigation of the most agricultural soils in the study area. The main crops in this area are rice, wheat, vegetables (*i.e.*, tomato, potato, eggplant, pepper and cucurbits), citrus, grape and mango. In addition, cucumber and pepper are the main vegetables cultivated in greenhouses

Sampling and analytical processing.

Sixty sites were selected for different upper horizon (0 - 30 cm) of soil samples which located in rural area and not close to possible industrial sites of pollution during October and November 2020. Soil samples were taken the major vegetable production fields over the Eastern Nile Delta (Table S1 and Fig. 4). For each sampling site, five subsamples of soil consisting of one center and four corners were pooled and homogenized to form a composite sample that covered approximately 20 × 30 m². The Global Positioning System (GPS) was used to determine the locations of samples on the spot (Fig. 4).

Soil characterization.

Soil samples were set in plastic bags and shifted to the soil laboratory that were air-dried for two weeks at room temperature (18 - 22°C). Then, the soil samples were sieved using a 2 mm aperture sieve and kept for more analyses. To estimate the particle-size distribution of investigated soils, the surface soil samples were conducted using the Pipette method according to Dewis and Freitas⁴³. The pH was measured in 1:2.5 soil in deionized water approximately 27°C to solution ratio using Beckman glass electrode pH meter⁴⁴. Electrical conductivity (EC) was detected in soil paste extract⁴⁴, while the soil organic matter was measured by the titration method based on the oxidation of organic matter by K₂Cr₂O₇⁴⁴. All selected soil samples were analyzed in triplicates and the averages were calculated by three values.

Digestion and estimation of trace metals.

For trace metal determinations, the soil samples were oven dried at 105°C overnight, sieved mechanically using a 0.5 mm sieve, homogenized and then crushed to 63 μm nylon mesh sieve according to the protocols described by Li et al.⁴⁵. Subsequently, 1g ± 0.01 of ground soil samples were weighed and digested in a Teflon bottles using of nitric acid (HNO₃) and perchloric acid (HClO₄) mixture by ratio of 4:1. Laterally, the samples were heated at 40°C for one hr. and augmented to 170°C for four hrs. till a clear solution was observed⁴⁶. After filtration with Whatman No.1 filter paper, the acidic solution is diluted with deionized water to a total volume of 50 ml. The total heavy metal concentrations (mg kg⁻¹ soil) were estimated using Thermo Scientific TM ICAPTM 7000 Plus Series ICP-OES⁴⁷. Ultimately, to confirm the correctness of estimation, a certified reference material (CRM 1570) was occupied according to the National Institute of Standards and Technology Standard. Triplicate analyzed samples were laid out to keep the quality of the dealings and the obtained results illustrated that all the estimated trace metals were analyzed with 98.2% recovery level.

Risk assessment of soil pollution.

The pollution indices of nine trace metals in the vegetable field soils from the Eastern Nile Delta was assessed by enrichment factor, geo-accumulation index, and contamination factor^{48,49}.

Enrichment Factor (EF).

The enrichment factor (EF) of trace metals is based on the measured metal standardization against a reference metal. The reference metal should be of natural origin in the study area and Fe was utilized as the normalization metal⁵⁰. The EF was calculated using Fe as the reference, according to the following equation:

$$EF = \frac{(C_n/C_{Fe})_{sample}}{(C_n/C_{Fe})_{background}} \quad \text{Equation 1}$$

Where (C_n/C_{Fe}) *sample* is the ratio of trace metal to iron concentration in the soil sample, and (C_n/C_{Fe}) *background* is the ratio between the background value of trace metal and the background value of iron.

The ecological risk based on the EF values were categorized into six groups, are listed in Table S2.

Geo-accumulation index (I_{geo}).

The geo-accumulation index (I_{geo}) was used to estimate the metal load enrichment in soil above the background level. It was calculated using the following equation that was offered by Muller⁵¹:

$$I_{geo} = \text{Log} \left[\frac{C_n}{1.5 \times B_n} \right] \quad \text{Equation 2}$$

Where C_n was the trace metal n concentration in soil and B_n is the background value of trace metal n (Cd = 0.3, Co = 19, Cr = 90, Cu = 45, Fe = 47200, Mn = 850, Ni = 68, Pb = 20 and Zn = 95 mg kg⁻¹). The constant 1.5 was applied to balance the probable natural differences in the values of background that may be attributed to various lithologic effects⁵².

Muller⁵¹ classifies the *Igeo* value into seven categories (Table S2).

Contamination factor (*CF*).

The contamination factor (*CF*) was computed using the following equation⁴⁸:

$$CF = \frac{C_n}{B_n} \quad \text{Equation 3}$$

Where C_n is the trace metal concentration in the soil and B_n is the value of metal background. Four categories that are shown in Table S1 were proposed by Hakanson⁴⁸ to classify the level of value of *CF*

Health risk assessment.

In the present study, risk assessment parameters and methods reported by the United States Environmental Protection Agency (USEPA) were used to assess the non-carcinogenic and carcinogenic health risk of trace metal in soils. These risks were calculated through ingestion (*ing*), dermal (*derm*) and inhalation (*inh*) routes for adults and children.

The average daily intakes (*ADIs*) for each metal were estimated using the following equations⁵³:

$$ADI_{ing} = \frac{C_s \times R_{ing} \times EF \times ED}{BW \times AT} \quad \text{Equation 4}$$

$$ADI_{derm} = \frac{C_s \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \quad \text{Equation 5}$$

$$ADI_{inh} = \frac{C_s \times R_{inh} \times EF \times ED}{PEF \times BW \times AT} \quad \text{Equation 6}$$

Where C_s is the trace metal concentration in the soil (mg kg⁻¹). The other exposure parameters and their symbols, values and units are presented in Table S3 in supplementary material⁵⁴.

The non-carcinogenic risks were evaluated using hazard quotient (*HQ*), hazard index (*HI*). The *HQ* value of each toxic metal for different exposure pathways is defined as Eq. (7)^{55,56}:

$$HQ = \frac{ADI}{RfD} \quad \text{Equation 7}$$

Where *RfD* (mg kg⁻¹ day⁻¹) is the reference dose of a trace metal that are listed in Table S4 in supplementary material.

The *HI* is the summation of multiple exposure pathways of *HQ* for each trace metal:

$$HI = \sum HQ \quad \text{Equation 8}$$

If the obtained *HQ* or *HI* values are less than 1, there is no non-carcinogenic health risk. If these values exceed 1, there may be significant risk to human health.

The carcinogenic risks (*CR*) of trace metals in soils were computed by Equation (9):

$$CR = \sum ADI \times SF \quad \text{Equation 9}$$

Where *SF* is the cancer slope factor of the trace metals. The values of *SF* are assumed in Table S3.

CR below 1×10^{-6} is considered that there is no significant effect to human body, *CR* between 1×10^{-6} and 1×10^{-4} is an acceptable range, and *CR* surpassing 1×10^{-4} is considered that there is a large potential carcinogenic risk.

Statistical analysis.

Descriptive statistics related to the trace metal concentrations and soil properties were examined to explore the data. The data was checked for the frequency distribution by Kolmogorov–Smirnov method. Pearson correlation analysis was performed to define the relationship among the soil variables. The cluster analysis was conducted to divide trace metals into different categories based on their sources in the soils of study.

Principal component analysis (PCA) was applied using factor extraction to identify the possible sources of nine trace metals in the study area. Components with eigenvalues higher than one unit after varimax rotation were taken^{57,58}. MSTAT-C, Microsoft Excel® (2013), and Number Cruncher Statistical System (NCSS) statistical software were used for the data analyses. Maps were prepared with ArcGIS (version 10).

Conclusions

This is the first study of risk assessment of the pollution of vegetable field soils by trace metals in Egypt. The results showed that the overall quality of vegetable fields in the Eastern Nile Delta is relative safety, although some samples impose serious pollution problems by Cd and Cr. The mean contents of Cu, Mn and Ni in soil samples were lower than their corresponding background concentrations, while the mean values of Cd, Co, Pb and Zn were exceeded their background values. The high concentrations of metals coupled with high variation can demonstrate that the anthropogenic activities may be their primary source in the study region. The determined enrichment factor, geo-accumulation index, and contamination factor revealed the study soil experienced low to moderate pollution, and the Cd and Cr pollution was very serious. The hazard index values from nine trace metals through the three exposure pathways for both adults and children were in acceptable range and far lower than the safe level, demonstrating that there was no a non-carcinogenic hazard for these age groups. The carcinogenic risk of Cd, Co, Ni and Pb metals were within the acceptable range for both adults and children, while Cr had significant carcinogenic health risk to children in the study area. Further studies are required to assess the health risk of trace metal intake from vegetables. The study highlights the importance of monitoring trace metals in vegetable field soils, enhancing soil management, decreasing the application of chemical fertilizers and pesticides, and irrigating with fresh water as excellent solutions for the long term to reduce further soil pollution in the vegetable field soils of the Eastern Nile Delta region.

Declarations

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Authors contributions

E. A. I. and E. M. S. conceptualize a research study. E. A. I. developed the design of the manuscript and prepared draft manuscript and figures. E. A. I. and E. M. S. designed the final manuscript structure, reviewed the scientific literature, and supervised the final version of the manuscript. The authors read and approved the final manuscript.

Additional information

Supplementary information to this article can be found online at

Competing Interests: The authors declare no competing interests.

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Figures

Dendrogram of Rows

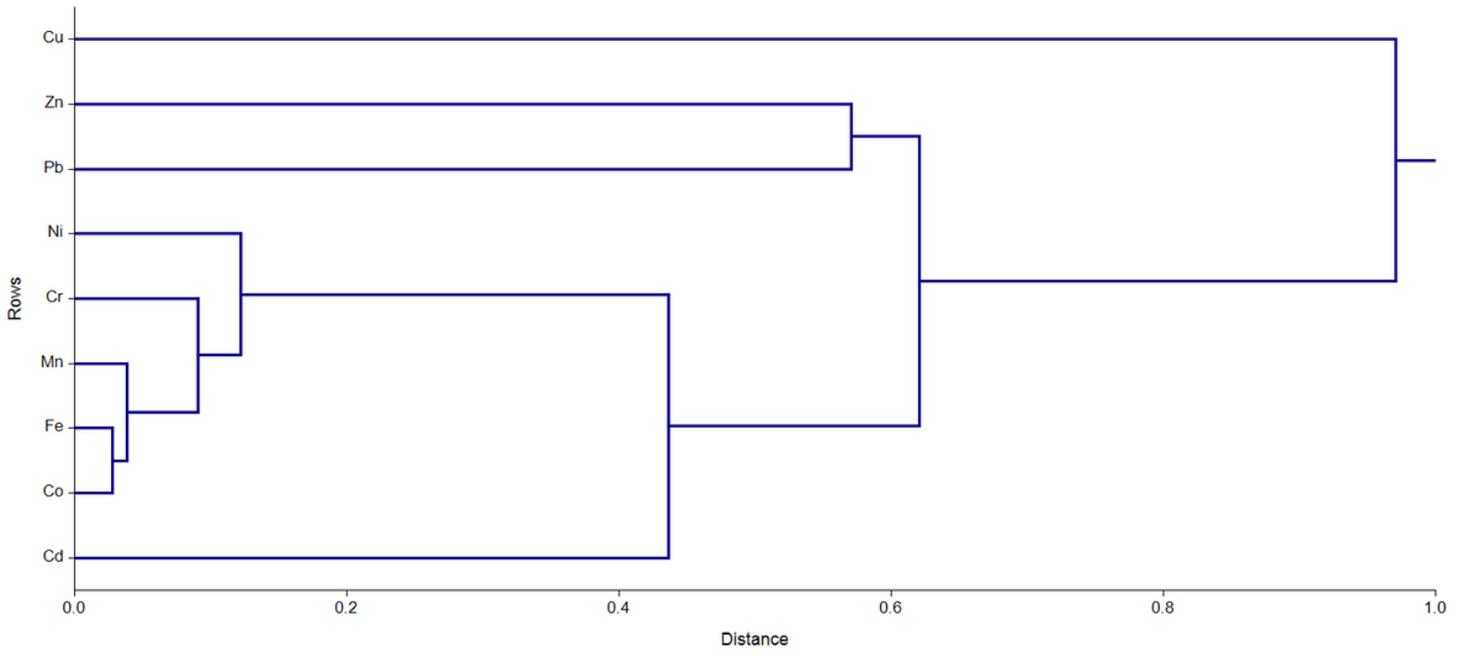


Figure 1

Hierarchical clustering of trace metals in vegetable soils in the Eastern Nile Delta.

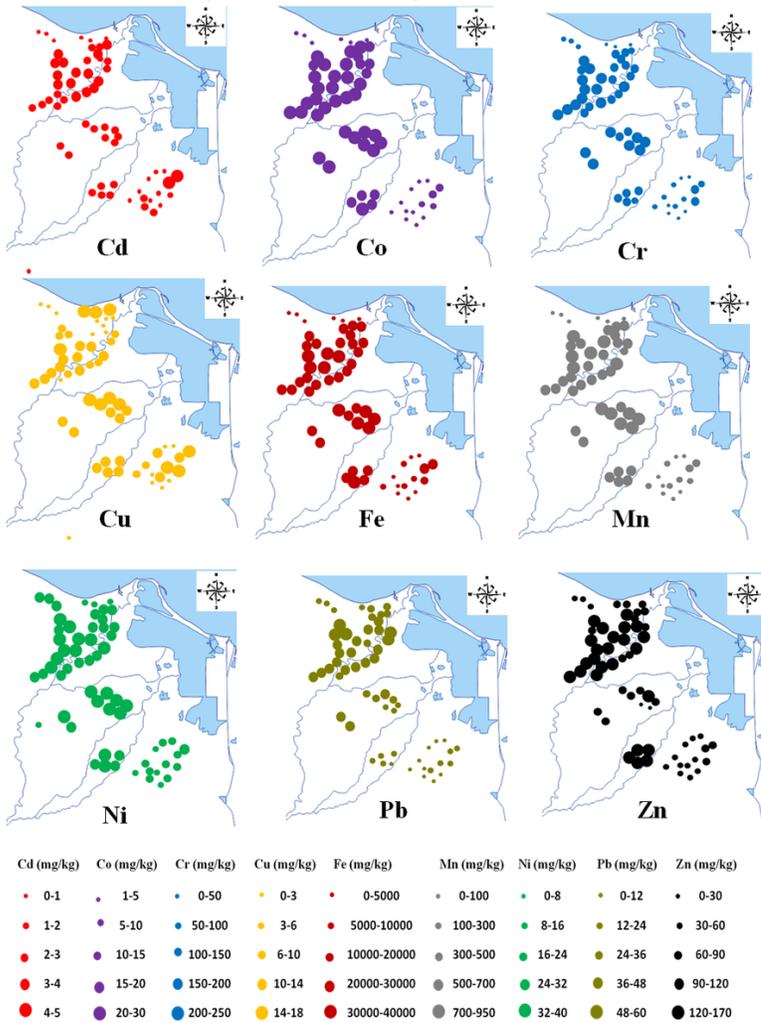


Figure 2

Spatial distribution maps of trace metals in the samples of vegetable field soils from the Eastern Nile Delta.

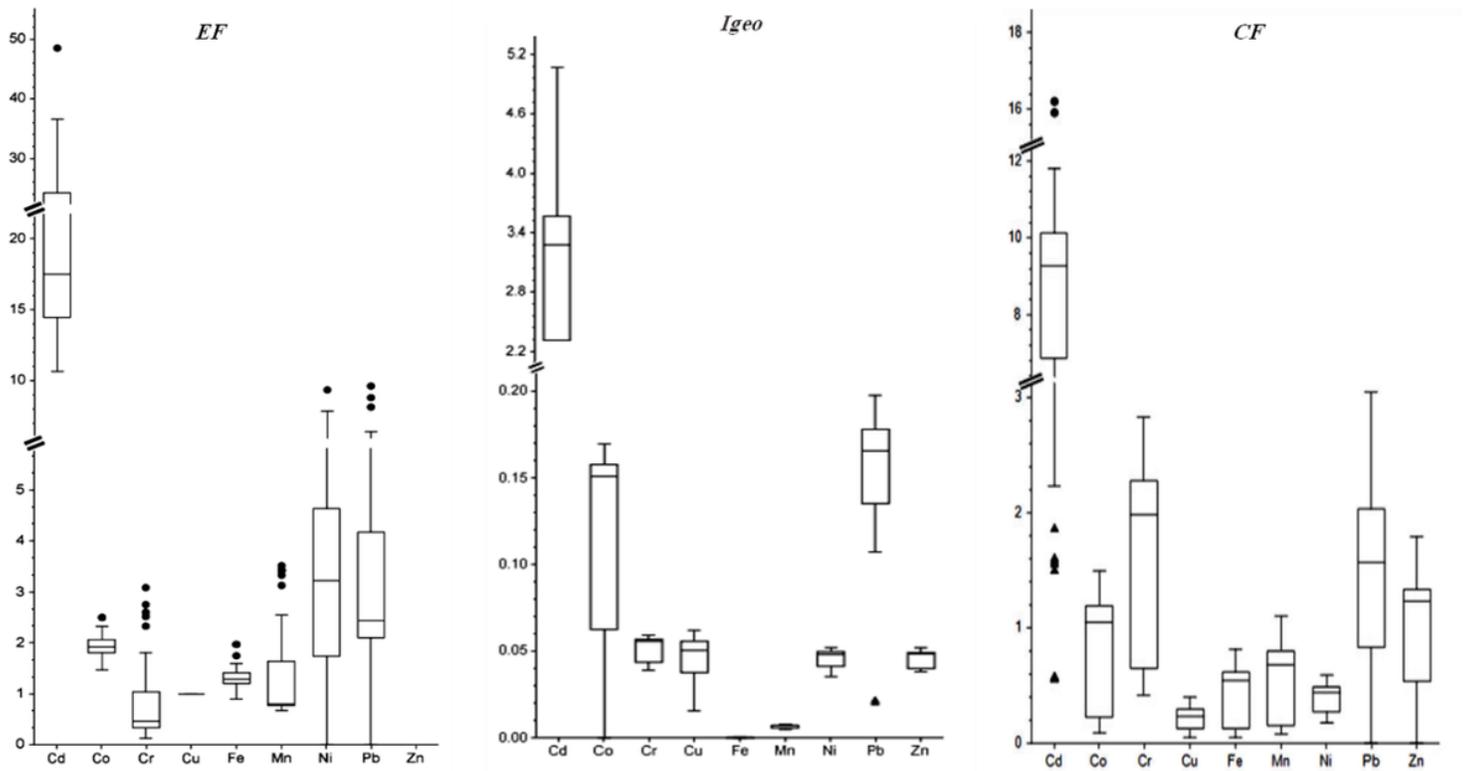


Figure 3

Box-whisker plots of the EF, Igeo and CF of trace metals in soils (the whisker shows the minimum and maximum values and the line of each plot is the median value).

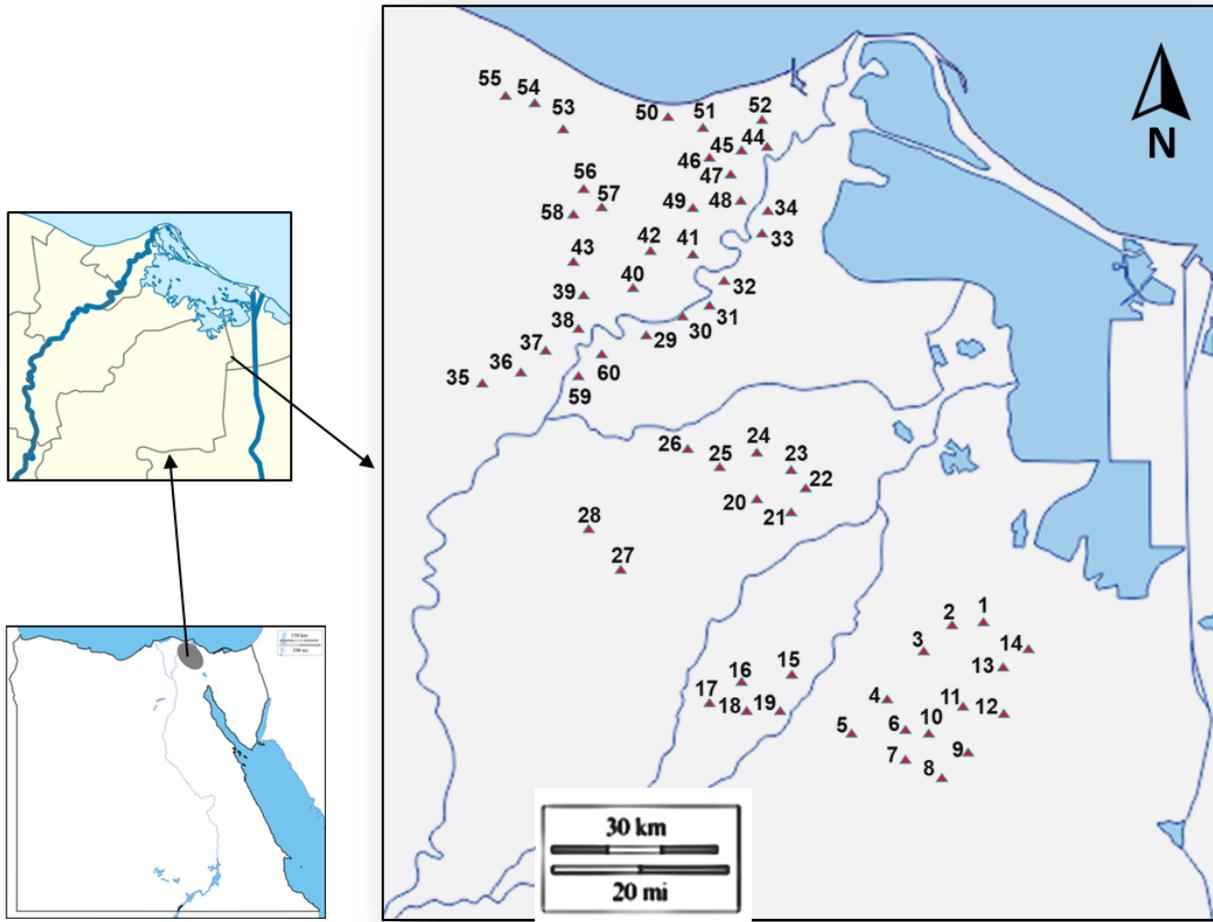


Figure 4

Map of study area and sampling sites of trace metals in vegetable field soils in the Eastern Nile Delta

Supplementary Files

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