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Spatial distribution of heavy metals content in soils of Amik Plain (Hatay, Turkey)

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Abstract The purpose of the study was to investigate the total and bio-available contents of heavy metals (Cd, Co, Ni, and Pb) and to determine their spatial variability in the Amik Plain, Turkey. Samples of surface and subsurface soil were collected at 132 sites in the research area. All of the total metal concentrations except Ni were considerably lower than their maximum allowable concentrations. Mean available Pb concentrations at both depths were above the permissible limits, while the other metal concentrations were within the proposed limits. Semivariograms of all the total metal contents, pH, and available Ni and Pb content were best fitted to spherical models, while available Cd and Co contents were best fitted to exponential models. Block kriging was used to interpolate values at unmeasured locations, generating maps of spatial variation for each heavy metal and pH.

Keywords Soils · Heavy metals · Geostatistics · Block kriging · Amik Plain

Introduction

Soils, the main part and the most endangered component of the terrestrial ecosystem, is a habitat for a great number of organisms and open to influences from a variety of pollutants arising from human activities (industrial, agricultural etc.). In this respect heavy metals are among the serious pollutants in soil due to their toxicity, persistence, and bio-accumulation (Morton-Bermea et al. 2002). Accumulation of heavy metals in soil not only reduces biological activity and lowers nutrient availability but also poses a serious threat to environmental and human health by entering into food chains and underground waters via the respective plant uptake and leaching process (Man-Zhi et al. 2006).

Accumulation of heavy metals in arable soils is important because of their potential transfer through crops to animals (feed crops) and to humans (food crops and vegetables; DeTemmerman et al. 2003). Applications of metal-contaminated sewage sludge, fertilizers, and animal manures can result in high concentrations of heavy metals in agricultural soils (Wu et al. 2009). Heavy metal concentrations and their spatial distributions can be used to assess the extent and origin of soil pollution. Free of human interference, the heavy metal contents of soil depend largely on the mineralogical composition of the parent materials and

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on the processes of weathering to which the parent materials have been subjected (Tack et al. 1997).

Geostatistics provides an advanced methodology which facilitates spatial interpolation and quantification of variability in soil variables measured and has become a useful tool for the study of spatial uncertainty and hazard assessment (McGrath et al. 2004; Robinson and Metternicht 2006). Geostatistical approach has been popularly applied to analyze spatial structure and spatial distribution of soil heavy metals (Imperato et al. 2003; Coşkun et al. 2006; Liu et al. 2006; Ahsan et al. 2009; Zhang et al. 2009; Wu et al. 2009). Kriging is one of the most precise estimators for spatial data analysis as it is unbiased and minimizes total uncertainty (Isaaks and Srivastava 1989).

Although the Amik Plain is one of the most important agricultural areas of Turkey, the spatial distribution of soil heavy metal concentrations still have been largely unknown. The objective of this study was to determine the total and plant-available concentrations of cadmium (Cd), cobalt (Co), nickel (Ni), and lead (Pb) and their interrelationships using kriging techniques in order to reveal their spatial distribution patterns as well as provide a basis for hazard assessment.

Material and methods

Study area

Amik Plain which covers about 75,000 ha of land is located in the Hatay province of Turkey (35°47′–36°24′ E; 35°48′–36°37′ N). The study region, one of the most important agricultural areas of Turkey, has a border with Syria and, surrounded by Reyhanlı, Kırıkhan, and Antakya city and Serinyol town. The study area has a typical Mediterranean climate with 1,124 mm annual precipitation and 18°C average annual temperature (Gün and Erdem 2003). Parent materials of the study area consist mostly of alluvium and lacustrines. Lacustrines are relatively flat and often have uniform properties. The alluvial soils formed by Orontes, Afrin, and Karasu rivers are the most productive soils. Lake Amik of ca. 53 km^2 in the northwest of the Amik Plain was drained into the Orontes River in order to increase the area of croplands (Kiliç et al. 2004, 2006).

Sampling and analyses

The study region was divided into grid squares of 2.5 × 2.5 km. A total of 264 soil samples were collected at intersections of the grids (total of 132 point) at depths of 0–20 and 20–40 cm. The geographic coordinates of sampling points in Universal Transverse Mercator System were determined with the help of global positioning system receiver (accuracy of ±5 m) in September 2003 (Fig. 1).

Soil samples were air-dried, passed through a 2 mm polyethylene sieve and analyzed for pH and heavy metals including Cd, Co, Ni, and Pb. Soil pH was measured in a 2.5:1 ratio of water to soil suspension using a glass pH electrode (Richards 1954). The total heavy metal concentrations were obtained by acid digestion according to Method 3050B (EPA 1996). The bio-available (available for plant uptake and microbial assimilation) fraction of the studied metals was extracted with DTPA (Lindsay and Norvell 1978). All heavy metal concentrations were determined by inductively coupled plasma atomic emission spectrometry (Varian liberty series II, axially viewed). All measurements were performed in triplicate. The analytical accuracy was acceptable since the relative standard deviation between the three parallel measurements was less than 5%. The accuracy of methods was also checked by analysis of high-purity certified standards, a reference material (Loam Soil C, Lot No. 707904).

Statistical methods

Descriptive statistical analysis (mean, minimum, maximum, median, coefficient of variation, skewness, kurtosis of heavy metal levels, and pH) was conducted as a first step towards an exploratory analysis of heavy metal data. Correlation analysis was used to assess the possible relationship among the heavy metals and pH. All statistical analyses were performed using SPSS 11.5.

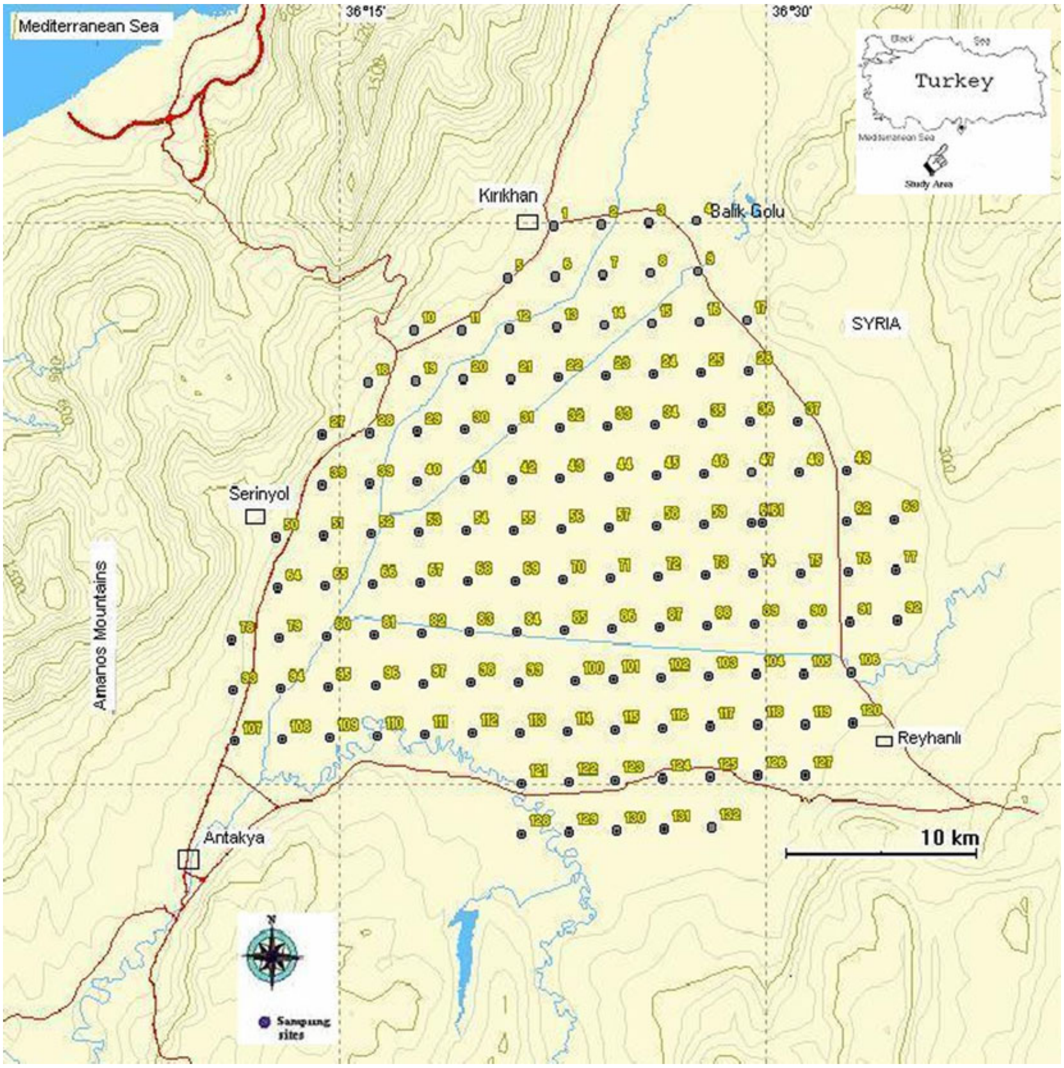

Fig. 1 Study area and sampling locations

Spatial prediction methods

Spatial variability in concentration of heavy metals has been determined using geostatistical methods. The experimental semivariograms were developed to reveal the spatial dependence of soil properties using the following equation (Lark 2000).

$$\gamma(h) = 1/2N(h)\sum\left[Z(xi) - Z(xi+h)\right]^2$$

where $\gamma(h)$ is the semivariance; $N(h)$ is the number of experimental pairs separated by a distance h; $Z(xi)$ is the measured sample value at point i and $Z(xi + h)$ is measured sample value at point $i + h$. From analysis of experimental variogram, a suitable model is fitted usually using weighted least square and such parameters as range, nugget and sill prior to the kriging procedure. All geostatistical analyses were carried out using GS$^+$ geostatistical software (version 7.0).

Selection for semivariograms model was carried out considering coefficient of determination (r^2) and scatter plots. The ratio of nugget variance (C_0) to sill variance ($C_0 + C$), which represents total spatial variation expressed in percentage, can be regarded as a criterion to classify the spatial dependence of soil variables (Cambardella et al. 1994; Chien et al. 1997). If the ratio was less than or equal to 25%, the variable was considered strongly spatially dependent (*S*); if the ratio was between 25–75%, the variable was considered moderately spatially dependent (*M*); and if the ratio was greater than 75%, the variable was considered weakly spatially dependent (*W*; Sun et al. 2003; Mehrjardi et al. 2008). If the slope of semivariogram was close to zero, regardless of nugget ratio, the variable was considered to be randomly distributed (*R*; Cambardella and Karlen 1999).

Results and discussion

Exploratory data analysis

Among the heavy metals and pH, the coefficients of variations (CV) were the highest for Ni and the lowest for pH in the soil layers (Table 1). Low CV values indicate a homogeneous distribution of soil variables, while high CV values indicate a non-homogenous distribution of variables in the study area. For example, the mean pH values in both soil depths were close to that of median values, whereas mean Ni values in both soil depths and fractions were fairly different from those of median values. In other words, the range of pH values was less variables than the range of Ni values (Table 1). Since pH values are on log scale of proton concentration in soil solution, it shows the lowest CV. There would be a much higher variability if soil acidity is expressed in terms of proton concentration directly (Sun et al. 2003).

Naturally elevated Ni contents are observed in soils over basic and volcanic rocks and especially in soils derived from serpentine rocks where Ni ranges from 770 to 7,375 mg kg^{-1} and under natural conditions, Ni toxicities are associated with serpentine or other Ni-rich soils (Kabata-Pendias and Mukherjee 2007). There is a wide area of serpentine rocks in the West boundary of the Amik Plain (Yıldız and Taptık 2003). In addition, it may probably be sediments with serpentine on Amik Plain. Nevertheless, naturally high Co contents are observed in soils over serpentine rocks (Kabata-Pendias and Mukherjee 2007). Natural Pb content in soils stems from parent materials. Its abundance in sediments is a function of clay fraction content, and thus, argillaceous sediments contain more Pb than sands, sandstones, and limestones. The total metal concentration in the study area occurs in sequence Ni >Co > Pb> Cd.

Table 1 Descriptive statistics for total and bio-available metal concentrations and pH of soils ($n = 132$)

Soil property	Depth (cm)	Min.	Max.	Mean	Median	SD	CV	Skew	Kurt
pH	0–20	7.20	8.75	7.93	7.94	0.26	3.28	–0.26	0.92
	20–40	7.17	8.39	7.93	7.96	0.22	2.78	–0.72	0.88
Total metal concentration									
Cd (mg/kg)	0–20	0.02	0.65	0.19	0.15	0.11	57.90	1.87	3.73
	20–40	0.02	0.65	0.16	0.14	0.09	56.25	2.23	7.29
Co (mg/kg)	0–20	0.40	74.00	20.42	14.45	13.32	65.23	1.76	2.70
	20–40	4.64	64.24	19.31	15.44	9.72	50.34	1.92	4.34
Ni (mg/kg)	0–20	46.60	1,305.73	273.57	183.28	225.89	82.57	2.50	6.91
	20–40	51.37	3,965.32	290.80	189.71	372.98	128.23	7.63	72.65
Pb (mg/kg)	0–20	0.60	20.94	5.56	4.35	4.12	74.10	2.13	4.29
	20–40	0.71	15.28	4.89	4.44	2.66	54.40	2.12	5.47
Bio-available metal concentration									
Cd (μg/kg)	0–20	0.89	64.83	20.81	18.05	12.84	61.70	1.21	1.66
	20–40	0.96	77.02	21.53	17.21	14.45	67.11	1.46	2.36
Co (μg/kg)	0–20	3.20	357.66	47.55	32.51	51.66	108.64	3.31	13.58
	20–40	5.26	156.95	34.28	26.81	25.84	75.38	2.29	6.58
Ni (mg/kg)	0–20	0.43	29.06	2.88	1.91	3.65	126.74	4.65	26.59
	20–40	0.54	23.97	2.86	1.73	3.31	115.73	3.56	15.59
Pb(mg/kg)	0–20	0.09	2.04	0.55	0.45	0.38	69.10	2.04	4.47
	20–40	0.10	2.22	0.56	0.45	0.40	71.43	2.22	5.48

SD standard deviation, *CV* coefficient of variation (%), *Skew* skewness, *Kurt* kurtosis, *Min.* minimum, *Max.* maximum

This results were compatible with outcomes by Dora et al. (2006).

DTPA metal concentrations of the soils may be considered to be the available fraction of the metal for microbial assimilation and plant uptake (Massas et al. 2009) that may readily enter food chain. Mean available Pb concentrations at both depths were above the permissible limits for bio-available concentrations in soils (0.2–2.0 mg kg^{-1}), as determined by DTPA extraction (Kaur and Rani 2006), while the other metals concentrations were within the proposed limits. Derici et al. (2002) reported that the available Cd concentrations in Amik Plain were determined higher than those in Mersin and Adana province. The available metal concentration occurs in the following sequence of Co > Cd > Ni > Pb. Our results were similar to those of Dora et al. (2006).

According to Kabata-Pendias and Mukherjee (2007), maximum allowable concentrations (MAC) for the total fraction of Cd, Co, Ni, and Pb in agricultural soils were 5 mg kg^{-1}, 50 mg kg^{-1}, 60 mg kg^{-1}, and 300 mg kg^{-1}, respectively when soil pH was greater than 6.0. Mean total Ni concentrations in both depths were four times higher than that of MAC. However, the mean total Cd, Co, and Pb contents were quite lower than those of MAC.

Geostatistical analyses

Since the probability distributions of the metal concentration data were heavily skewed, the experimental semivariograms were developed using transformed data except for pH to stabilize variance (Goovaerts 1999). Logarithmic transformations of all the metal concentration data except for bio-available Cd resulted in smaller skewnesses and kurtosis. Square root transformations for bio-available Cd showed similar trend. The pH values presented nearly normal distribution, thus transformation was not carried out for pH prior to further analysis. The summary of the semivariogram models for heavy metals and pH is given in Table 2 and in Fig. 2. In order to understand the distribution patterns of the four heavy metals and pH, block kriging interpolation was used to obtain the filled contours maps (Figs. 3, 4, and 5).

Isotropic variogram models were fitted in all the cases. Spherical model was defined for all the

Table 2 Best fitted semivariogram models and model parameters for heavy metals in soils (n = 132)

Soil property	Depth (cm)	Class, model	Range (m)	Nugget (C_0)	Sill ($C_0 + C$)	($C_0/C_0 + C$)	r^2
pH	0–20	M, spherical	14,830	0.035	0.071	0.492	0.943
	20–40	M, spherical	20,710	0.024	0.052	0.461	0.967
Total metal concentration							
Cd (mg/kg)	0–20	M, spherical	58,540	0.136	0.435	31.3	0.969
	20–40	M, spherical	17,560	0.098	0.303	32.3	0.831
Co (mg/kg)	0–20	S, spherical	48,100	0.156	0.657	23.7	0.971
	20–40	S, spherical	61,100	0.050	0.410	12.2	0.893
Ni (mg/kg)	0–20	S, spherical	61,100	0.047	0.973	4.8	0.897
	20–40	S, spherical	61,100	0.040	0.975	4.1	0.883
Pb (mg/kg)	0–20	S, spherical	61,100	0.092	0.886	10.4	0.986
	20–40	S, spherical	61,100	0.080	0.525	15.2	0.871
Bio-available metal concentration							
Cd (µg/kg)	0–20	M, exponential	18,690	0.959	2.074	46.2	0.829
	20–40	M, exponential	16,170	0.874	2.373	36.8	0.851
Co (µg/kg)	0–20	M, exponential	183,300	0.483	1.318	36.6	0.897
	20–40	M, exponential	122,070	0.304	0.608	50.0	0.938
Ni (mg/kg)	0–20	M, spherical	11,280	0.185	0.538	34.4	0.622
	20–40	M, spherical	10,080	0.147	0.560	26.3	0.541
Pb (mg/kg)	0–20	M, spherical	53,310	0.211	0.598	35.3	0.988
	20–40	M, spherical	61,100	0.190	0.717	26.5	0.973

S strong spatial dependence, *M* moderate spatial dependence

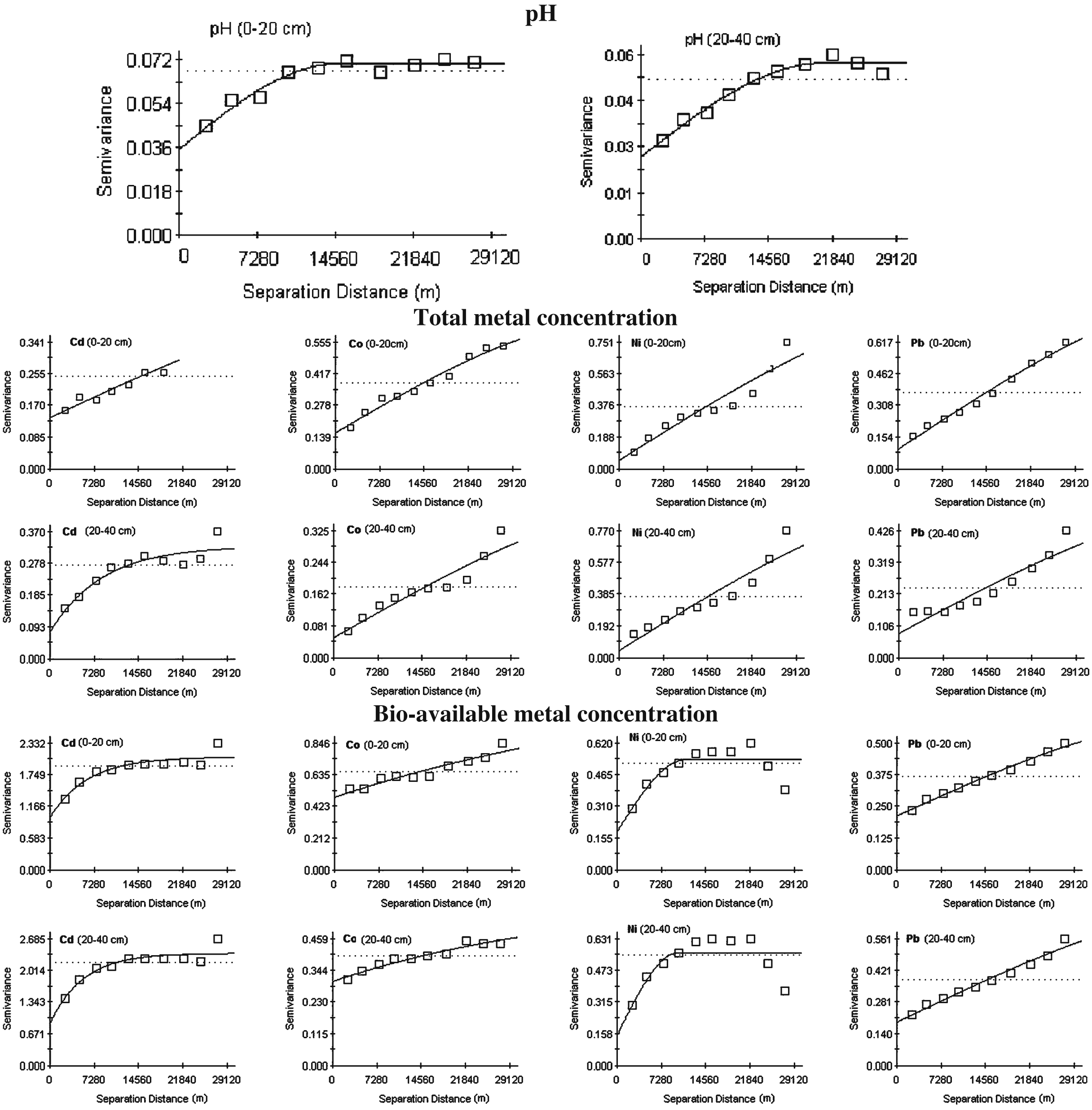

Fig. 2 Experimental semivariograms of pH and soil heavy metals

heavy metals and pH with the exception of available fraction of Cd and Co which were defined by exponential model for both depths. The spatial variability of soil properties may be affected by intrinsic (natural factors, such as soil parent materials) and extrinsic factors (anthropogenic factors, such as agricultural practices). Usually, strong spatial dependence of soil properties can be attributed to intrinsic factors, and weak spatial dependence can be attributed to extrinsic factors (Cambardella et al. 1994; Wu et al. 2009). In this study, the total contents of Co, Ni, and Pb exhibited strong spatial autocorrelations (ratios ranging from 4.1% to 23.7%). This case suggested that

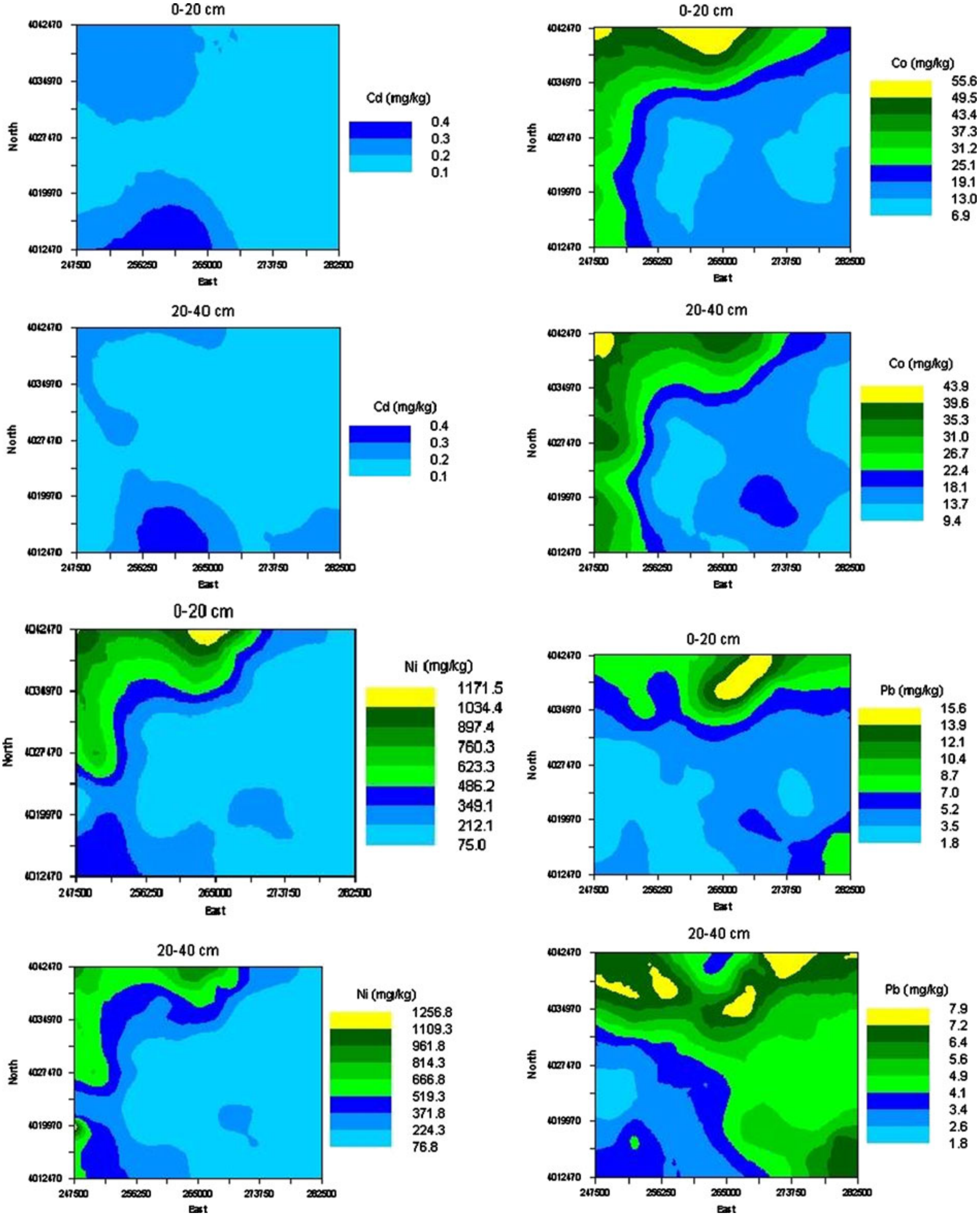

Fig. 3 Spatial distribution maps of soil total metal concentrations in the study area

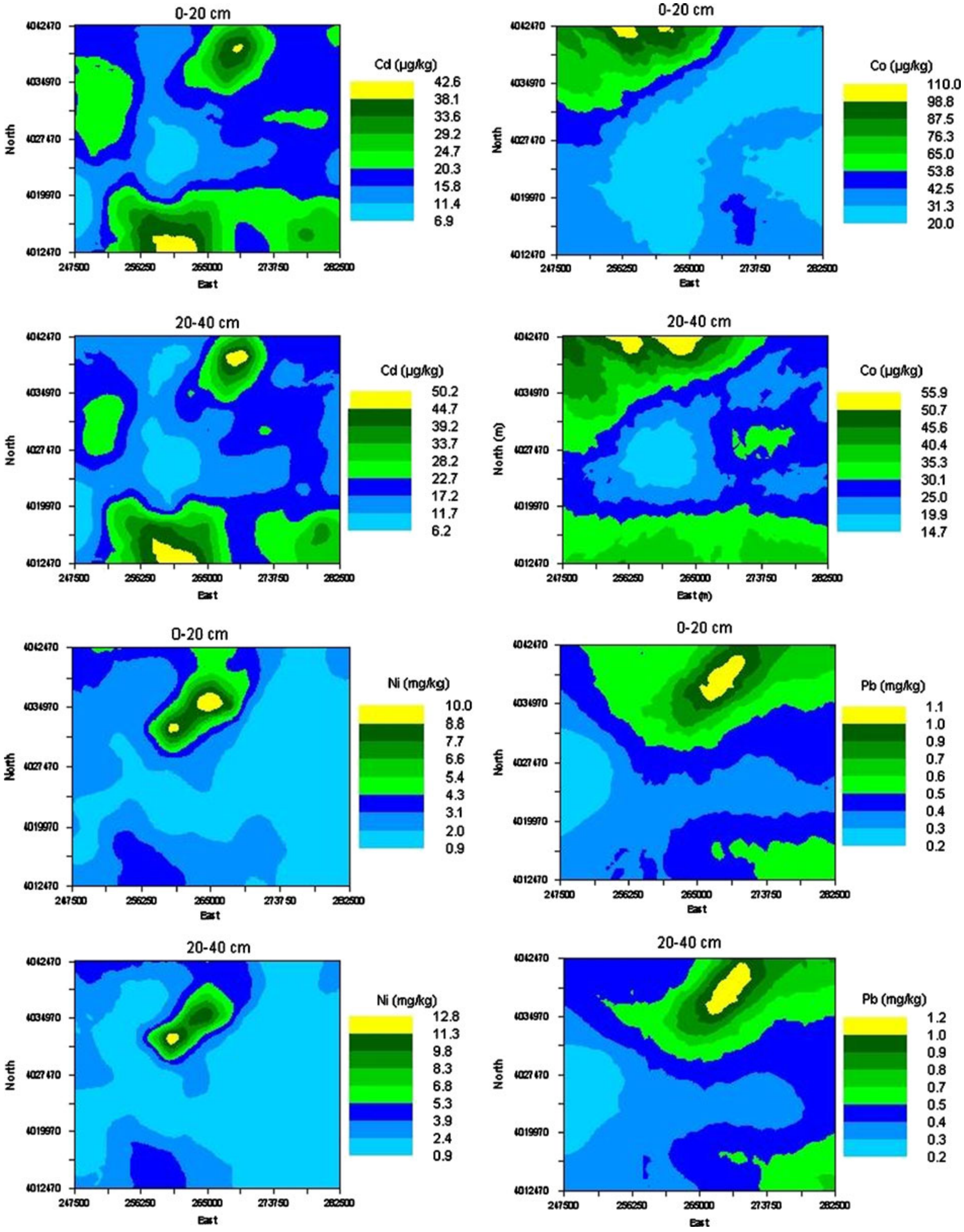

Fig. 4 Spatial distribution maps of soil available metal concentrations in the study area

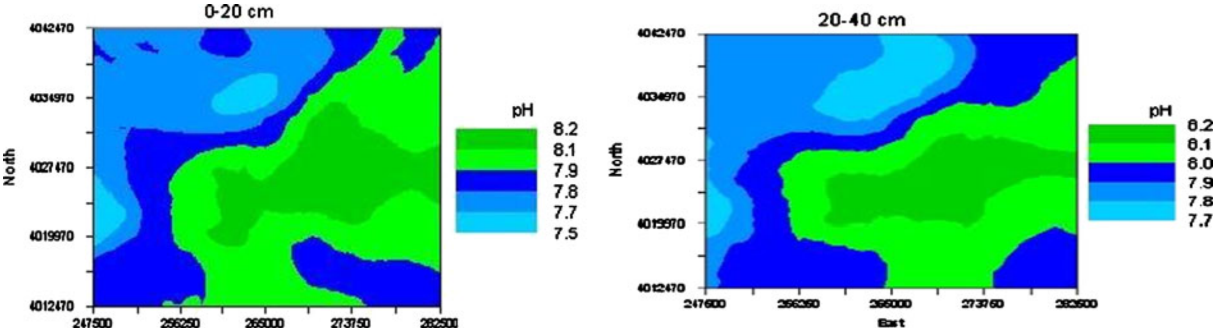

Fig. 5 Spatial distribution maps of soil pH

their spatial variability was affected by natural factors, whereas pH values and the total concentration of Cd presented moderate autocorrelation (ratio = 31.3%) indicating its spatial variability was affected by natural factors as well as human activities, especially P-fertilization since one of the major sources of Cd contamination in soils is also P-fertilizers (Kabata-Pendias and Mukherjee 2007). The pH values showed a moderate autocorrelation. The pH values may be affected by carbonate contents of the soils as most of the soils in the study area have high carbonate contents (Kiliç et al. 2004, 2006). pH is positively correlated with carbonate content of the soil. The available contents of all the heavy metals also exhibited moderate spatial autocorrelations (ranging from 26.3% to 50.0%; Table 2).

The ranges of semivariograms, which indicate the maximum distance of spatial correlation for heavy metals and pH, ranged from 10 km for available Ni content to 183 km for available Co content. The highest total Cd values were observed in the south part of the study area, whereas the highest total Co content was in the north and northwest sections of the area. The total Ni values were the highest in the northwest and the lowest in the east of the study area. The north part of the region shows higher total Pb levels than southwest part. In generally, this trend was observed at the two soil layers (Fig. 3). pH values in the soil depths were higher in the southeast than in northwest (Fig. 5).

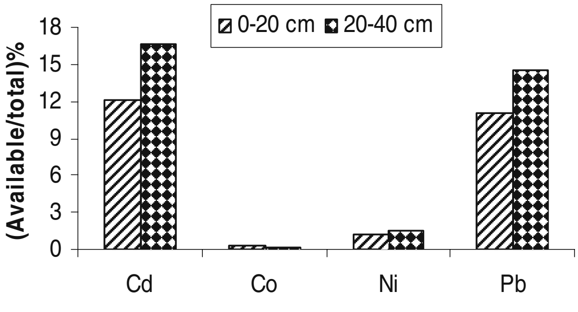

Fig. 6 Mean available/total concentration ratio (%) of the heavy metals at depths of 0–20 and 20–40 cm

To compare the availability of metals between the soil depths, the available concentrations-to-total concentration ratio (%) was calculated for each metal and sampling point. The mean values of this ratio for all the metals are presented in Fig. 6. The availability of the heavy metals in subsoil was slightly higher than that of topsoil. The availability of Cd and Pb was extremely higher than those of Co and Ni (Fig. 6).

Conclusions

It can be concluded that the kriging interpolation techniques can be successfully used to investigate spatial variation of heavy metals and pH in the Amik Plain soils. Since the ranges of semivariograms which indicate the maximum distances of spatial variability of soil properties were of very large distances (ranged from 10 to 183 km), sampling distance (2.5 km) is adequately suitable to prepare the distribution maps of the heavy metals. Therefore, the distribution maps produced for soil heavy metals in this research may be used confidently for the environmental and agricultural purposes. The large distances of the range of influence may show the importance of sampling the whole study area in a future research. In fact, sampling distance may be larger than 2.5 km and optimum number of soil samples may be lower than that of this work in further research in the same area.

The parent material and inorganic fertilizers were mainly responsible for the distribution of the metal contents in the study area since there were no industrial activities or heavy traffic in and around Amik Plain. The total Cd, Co, and Pb contents were quite lower and Ni content was four times higher than those of MAC. The available heavy metal contents are more important than total metal contents for the environmental health and agricultural production. As mentioned before, among all the heavy metals, the availability of Cd and Pb were higher (∼14.3% of the total) than those of Co and Ni. This indicated that these elements are potentially bio-available and may leach through the soil. Only mean available Pb concentrations at both depths were above the permissible limits for bio-available concentrations in soil. However, the total Pb contents are extremely lower than MAC. On the other hand, in spite of the total high Ni content, the available Ni content was quite low because Ni in soil is slightly mobile and occurs mainly in the residual fraction. Consequently, none of the four heavy metals had high risk for environmental pollution and agricultural production in the study area. But, in the future bio-available Cd content may increase in soils because P-fertilizers have been applied in large quantities to increase the agricultural production.

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