Eurasian J Soil Sci 2019, 8 (3) 249 - 256

Eurasian Journal of Soil Science

Journal homepage : http://ejss.fesss.org

Determination of heavy metal risk and their enrichment factor in intensive cultivated soils of Tokat Province

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Article Info

Received : 09.12.2018
Accepted : 15.05.2019

Abstract

Heavy metal contamination has caused serious environmental and health-related problems around the world. This research was conducted in arable lands of some basins located on Tokat province. The aim of this present study was to determine I-) some physico-chemical properties of soils, ii-) to find heavy metal (HM) content and their enrichment factor (EF) and iii-) to detect relationship between some physico-chemical properties and HM concentration. To identify the concentrations and sources of heavy metals, 280 soil samples (0-20 cm) were collected from the study area. Subsequently, in order to evaluate natural or anthropogenic sources of heavy metal content and their EF in agricultural fields, the concentrations of some HMs (Cd, Co, Cu, Cr, Ni, Pb and Zn) and some physico-chemical properties of soil samples were analyzed. The results showed that mostly the concentration of Ni followed by Cr exceeded their threshold levels. The local pollutions from Ni and Cr were attributed to the natural influences (particularly due to parent material). The concentrations of the other HMs are relatively lower than the critical values. The mean values of the HMs contents arranged in the following decreasing order: Ni>Cr>Cu>Zn>Co>Pb>Cd in the studied soil sample. In addition, it was found significantly positive relation between Pb and OM while the same relation was also found clay content and Cd and Pb. On the other hand, according to EF of HMs in total soil samples, Cd, Ni and Cr have found 16%, 10% and 6% soil samples as moderate enrichment class, whereas 55% and 1% of the total soil samples were determined significant enrichment class in terms of Cd and Ni elements. Besides, all other HM elements did not exceed minimal enrichment level. However, in some regions of the study area, the Cu, Cd and Pb contents were also slightly raised, this case possibly stems from anthropogenic effects such as excessive P fertilization, field traffic and pesticide using.

Keywords: Heavy metal risk, enrichment factor, micro basin, Tokat.

Introduction

The heavy metal pollution in agricultural lands has been very important in recent years, especially due to the detrimental effects on food safety and ecosystem. The source of this pollution in agricultural soils can be geogenic and/or anthropogenic. Geogenic pollution is entirely due to the composition of the main material that forms the soil. Anthropogenic pollution may be caused by excessive use of fertilizers and pesticides in agricultural areas, use of fossil fuels, mining activities, rapid population growth and related urbanization, uncontrolled wastewater discharge, atmospheric accumulation, traffic density and increased industrial activities (Manta et al., 2002; Borůvka et al., 2005; Sivry et al., 2008; Zhang et al., 2009; Meena et al., 2011, Bilge and Çimrin, 2013).

Heavy metals, which are added to the environment by natural and artificial means, are defined as dangerous pollutants because they easily accumulate and form complex structures in the soil. These heavy metals cause

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e-ISSN: 2147-4249 DOI: 10.18393/ejss.567357

decrease in microbial activity, soil fertility, biodiversity and yield, and can also cause poisoning in animals and humans through the food chain (Karaca, 2001; Oliveira et al., 2006; Yang et al., 2006; D'Ascoli et al., 2009; Peralta-Videa, 2009; Mudgal et al., 2010; Yadav, 2010; Kaplan et al., 2011; Jaishankar et al., 2014; Liu et al., 2017.). Some physical and chemical properties of soils such as cation exchange capacity, pH and organic matter are effective in the accumulation of these metals in soil. Especially heavy clay soils can absorb heavy metals in their bodies due to their high cation exchange capacity. Also in soils with high organic matter content, heavy metals are absorbed more and low solubility compounds occur (Bakış and Bilgin, 1998).

Tokat Province, selected area for this reseach, has been shown to be an important settlement center throughout its history with the advantage of being established on the fertile valley of Yeşilırmak. The fertile plains where all kinds of agriculture can be made are distributed all over the province. The most important of them are Kazova, Omala Plain, Turhal Plain, Niksar Plain, Erbaa Plain, Artova and Zile Plain. All kinds of fruits, vegetables and sunflowers, especially cereals, sugar beet and tobacco are grown in these plains. Approximately 16.5% of the province is composed of meadow and pasture areas and sparse plant areas, while 31.0% of them constitute forest areas. The agricultural activities area covers 38.2%. In addition some researcher performed about heavy metal concentration in arable lands such as Arda et al. (2015), in their study, took 42 soil samples from the Ipsala District and its villages and stated that the nickel concentrations determined in the Ipsala Region were significantly higher than the limit values (0.25-109.5 mg/kg). Moreover, Metin (2010) indicated that in the study it was determined the amount of trace elements and heavy metals in the agricultural lands of the Aluvial, Colluvial and Vertisol group in the western part of the Bursa plain, there was no pollution exceeding the limit values in terms of Cd, Co, Cr, Ni and Pb in the studied agricultural soils. Akyıldız and Karataş (2018) reported also that in the soil samples taken from Adana city center and its surrounding areas, Fe, Mn and Pb were found to be below the standards and Cu, Hg, Co, Cd and their elements were found to be above a few sample standards. In the analysis results of Ni, As, Cr and Al, samples were mostly above the standards. It is stated that the parameters that cause pollution are generally caused by environmental factors, and that the geological structure in the region may contribute in the increase of Cr and Ni elements.

In the study, which investigated the heavy metal contents and pollution characteristics in the water and sediments of the Khoshk River in southwestern Iran, it was stated that the amount of heavy metal in the sediments decreased as Mn>Cr>Pb>Ni>Zn>Cu>Cd respectively. Based on the enrichment factor and geoaccumulation index values, it was determined that the sediments were filled with Cr, Zn, Pb, Cu and Cd. According to the results obtained from the basic component analysis with sediment samples, it was stated that the high Ni concentration is related to the composition of the main rocks, whereas Cr, Zn, Pb, Cd and Cu values may be due to anthropogenic activities (Salati and Moore, 2010).

In order to determine the amount of Cd, Co, Cu, Ni, Pb, Zn in the Bafra deltaic plain and to analyze the spatial distribution of the heavy metal, 108 soil samples were taken from 0-20 cm depth from an area of approximately 100 thousand ha. In order to reveal the source of heavy metal pollution (natural or anthropogenic), the enrichment factor was calculated. The highest enrichment factor was found for Cd (12.826), while the smaller enrichment factor values for Pb, Ni, Co and Cu were calculated. The concentrations of Cd, Cu and Zn in some regions of the study area were determined to be slightly higher due to the high percentage of phosphorus fertilizers and intensive agricultural applications. It is stated that the values exceeding the criterium for Ni are due to the high content of this element in the main material (Kızılkaya et al., 2011).

The aim of this present study conducted in arable lands of some basins located on Tokat province was to determine i) some physico-chemical properties of soils, ii) to find heavy metal (HM) content and their enrichment factor (EF) and iii) to detect relationship between some physico-chemical properties and HM concentration.

Material and Methods

Field description

This study was carried out in Tokat province in the Central Black Sea region of Turkey. The Province of Tokat is coordinated between 4200000-4520000 North and 210000-360000 East (WGS84, UTM-37 Zone m) (Figure 1). Total area of the Tokat is about 10272.58 km^2. However, arable land selected for this study covers approximately 38.2% of the total area. On the other hand, 47.5% of the study area is covered by forest and pasture lands.

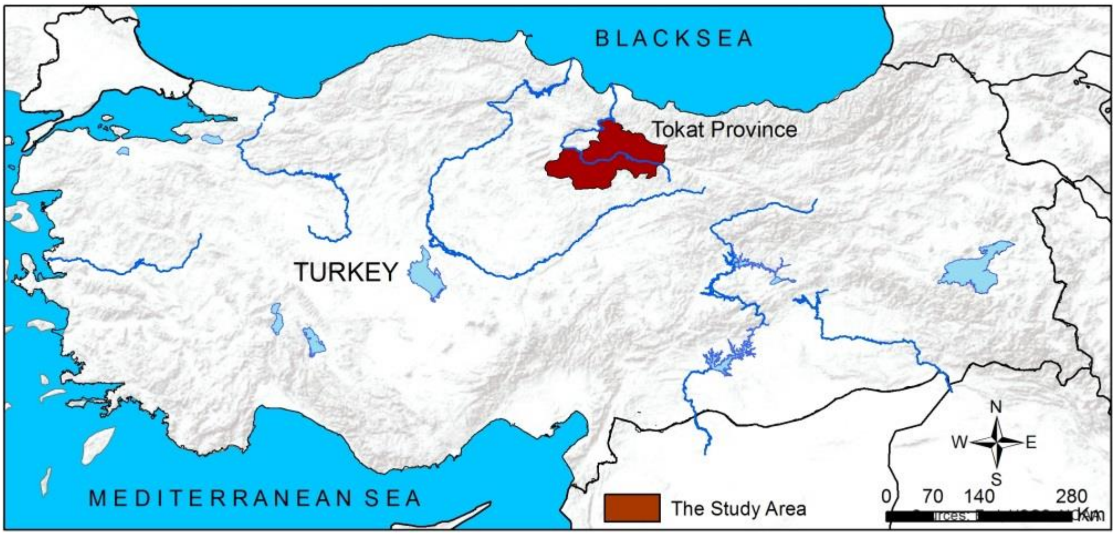

Figure 1. Location map of the study area

The climate can be described as sub-humid and according to long term meteorological data (1974-2017), average annual precipitation and temperature of the study area are 431.4 mm and 12.6 ºC, respectively. The study area lies at an elevation above the sea level from 75 to 2415 m. The region has topographically very heterogeneous topographic features such as hilly, rolling, flat, etc., but particularly hilly and rolling physiographic units are common in the study area, only 12.0% of the total area is almost flat and gentle slope and about 22.5% of it is less than 10% slope degree (Figure 2). Most of the total area corresponding with 796200.9 ha has more than 20% slope degree.

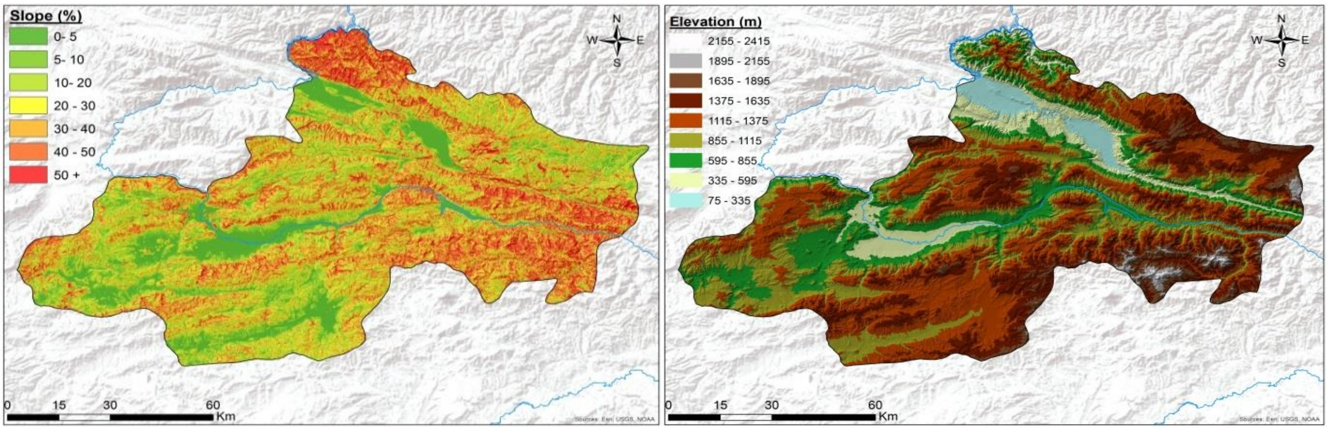

Figure 2. Elevation and slope maps of the Tokat Province

Soil Sampling and analysis

Field study was conducted in 2017. In total 280 soil samples classified as mostly alluvial, brown, brown forest, non-calcareous forest, chestnut and reddish chestnut great soil group were taken from soil surface (0-20 cm) in agricultural lands (Figure 3). The sampling was carried out after harvest in the autumn and before start of the next cropping season in order to avoid the influence of agricultural practices during the crop growing season, i.e. fertilization. In addition, their coordinates were recorded using global positioning system (GPS) tool. Samples were air-dried and sieved through a 2 mm sieve to be prepared for analyses. Soil requirements for organic farming including soil physico-chemical properties, heavy metal concentration were determined based on literatures. Table 1 shows the selected analytical protocols.

Table 1. Protocol measurements for some soil physical and chemical properties

Parameters	Unit	Protocol	Reference
Texture (Clay, Silt and Sand)	%	hydrometer method	Bouyoucos (1951)
pH	1:1	(w:v) soil-water suspension	Soil Survey Laboratory (1992)
EC	dSm^{-1}	(w:v) soil-water suspension	Soil Survey Laboratory (1992)
$CaCO_3$	%	Scheibler calcimeter	Soil Survey Staff (1993)
Organic Matter	%	Walkley-Black method	Soil Survey Laboratory (1992)
Total heavy metal (Cu,Cd,Cr,Pb,Co,Ni,Zn)	mg kg^{-1}	According to EPA 3051 sing ICP-OES detection	Kloke (1980)

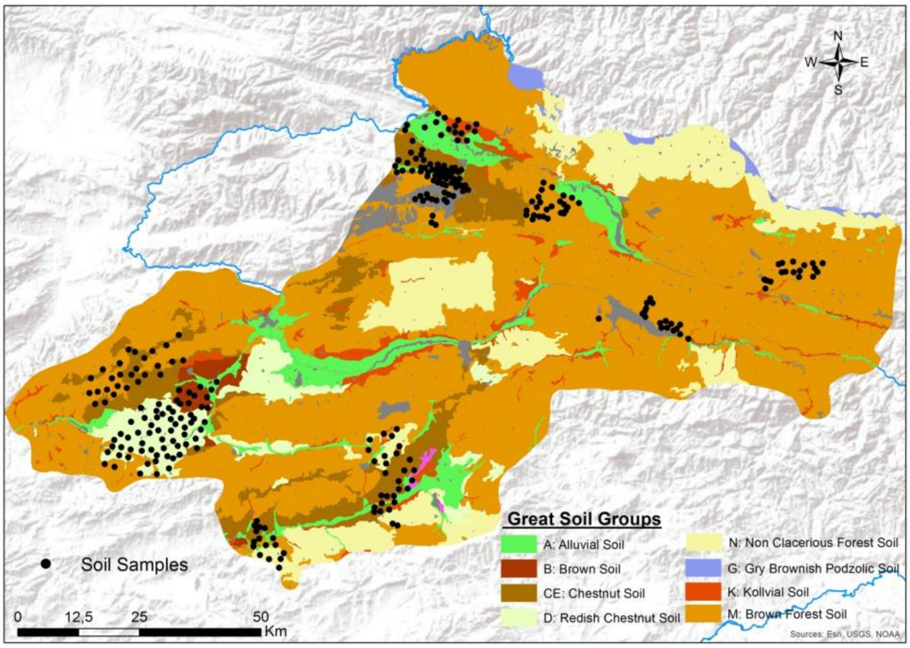

Figure 3. Soil samples pattern in great soil group map of the study area

Some physico-chemical characteristics of soil such as the organic matter, pH, and lime contents, and the particle size fractions are of great importance in the heavy metal toxicity of soils. The calculation of the enrichment factors (EF) for the heavy metals was made using an equation suggested by Sposito (1989) and Agbenin (2002).

$$EF= (HM_{soil}) / (HM_{earth})$$

where HM_{soil} is the total heavy metal concentration in the soil sample, and HM_{earth} is the mean heavy metal concentration in the earth's crust, which is 0.11 mg kg^{-1} for Cd, 50 for Cu, 100 for Cr, 20 for Co, 80 for Ni, 14 for Pb, and 75 mg kg^{-1} for Zn (Sposito, 1989). Based on the EF value, five categories of pollution were distinguished by Sutherland (2000): the absence of enrichment (<2), moderate enrichment (2–5), high enrichment (5-20), very high enrichment (20–40), and extremely high enrichment (>40).

Results and Discussion

Soil physico-chemical properties and heavy metals

The physico-chemical characteristic selected in this study showed changefulness as a result of dynamic interactions among natural environmental factors, including the degree of soil formation, leaching process, and agricultural activities such as tillage systems or fertilization (Dengiz et al., 2015). The descriptive statistical parameters such as mean, maximum, minimum, and coefficient~~s~~ of variation (CV) of the some basic physico-chemical properties related to 280 soil samples taken from surface (0-20 cm) of the crop lands in 21 micro catchments of the Tokat province were given in Table 2. In order to determine variability of some physico-chemical soil properties, many researchers offer to investigate coefficient of variation (CV). According to CV values, it was classified as low (<15%), medium (15-35%) and moderate (> 35%) (Mallants et al., 1996). In this case, variables of sand, silt of physico-chemical soil properties and Zn have moderate CV. On the other hand, the variables of clay, EC, $CaCO_3$ content and OM of soil physico-chemical properties and all HM (except for Zn) had a high level of variability. In addition to that, the values of pH in soil samples have low variability and ranged from moderately acid to slightly alkaline (5.22 and 8.07), whereas electrical conductivity had a mean value of 0.63 dS m^{-1}. The mean values of organic matter and $CaCO_3$ content (%) were 2.04 and 11.34. Table 2 shows also HM status of the soil samples.

As for the concentrations of heavy metals in the surface soils given in Table 2, it was determined that variables of heavy metal concentration of soils had a high level of variability. In the soils studied, the concentrations of Cu amounted to 6.83–106.11; Cr, 0.00-377.44; Pb, 0.00-19.80; Co, 0.00-27.55; and Ni, 2.79-521.92 mg kg^{-1}. The heavy metal contents such as Cu, Ni and Cr were higher than those given in Table 2. On the other hand, Pb, Co, Cd and Zn concentration not exceeded their permissible threshold level in all of the soil samples. While 51% of the soil samples was found Ni concentration exceeded its maximum permissible value and illustrated its grouping in representative micro catchments (Figure 4), Cr concentration passed maximum level of permission in 15% of soil samples. Furthermore, only in one sample an elevated Cu and Pb content was found.

Table 2. Descriptive statistical analysis of physico-chemical properties and heavy metal of soil samples

Parameters	Mean	SD	*CV	Variance	Min.	Max.	**Skewness	Kurtosis
Physico-chemical properties								
Sand, %	42.57	11.66	27.38	135.88	11.11	87.05	0.39	0.83
Clay, %	27.59	10.28	37.25	105.60	4.91	52.93	0.33	-0.22
Silt, %	29.84	8.01	26.83	64.12	7.73	67.27	1.29	3.42
pH, 1:2.5	7.45	0.46	6.19	0.21	5.22	8.07	-2.93	9.49
EC, dS m^{-1}	0.63	0.31	48.87	0.09	0.00	2.83	2.20	10.48
$CaCO_3$, %	11.34	9.41	82.99	88.56	0.00	53.05	0.90	0.95
OM, %	2.04	0.87	42.78	0.76	0.74	5.88	1.66	3.82
Heavy metal								
Cu (0-100)#	36.55	15.64	42.79	244.60	6.83	106.11	1.14	2.95
Cd (0-3) #	0.56	0.47	83.35	0.22	0.00	2.37	0.59	0.29
Cr (0-100) #	58.54	50.66	86.53	2566.18	0.00	377.44	2.61	9.68
Pb (0-100) #	6.93	3.70	53.44	13.71	0.00	19.80	0.36	-0.05
Co (0-50) #	8.63	5.59	64.71	31.22	0.00	27.55	0.80	0.61
Ni (0-50) #	67.04	67.86	101.22	4605.17	2.79	521.92	3.30	14.11
Zn (0-300) #	36.14	11.85	32.79	140.46	3.73	76.69	0.18	0.18

SD: Standard deviation, Min.: Minimum, Max.: Maximum, n: sample number, EC: Electric conductivity, OM: Organic matter.
*CV (Coefficient of Variation), **skewness:< │∓0.5 │= Normal distribution, 0.5- 1.0 = Application of character changing for dataset, and > 1,0 → application of Logarithmic change,
#: Maximum permissible concentration (mg kg^{-1})

According to results, micro catchments coded as 2, 3, 4, 11, 8, 12, 13, 14, 16, located around Zile, Sulusaray, Niksar, and Artova Districts were determined accumulation of Ni concentration which exceeded maximum permissible concentration (MPC) in surface soil soils of the northern east and southern west parts of the Tokat province area. This relatively high Ni concentration level is not related with industrial or other anthropogenic pollution. It can be said natural case namely; it appears to be associated with the properties of the parent (volcanic) rock. As Chen et al. (2005) and Kızılkaya et al. (2011) reported, the Ni concentration in volcanic rocks is 20–40 times greater as compared to other ones. All the heavy metal concentrations determined in the soils of the test plots were lower than the threshold ones. In addition to that, high Cu heavy metal content was found in one micro catchments coded as 8 located around Niksar District. This fact is likely related to the wide application of pesticides containing copper. In addition, in some micro catchments coded as 2, 3, 4 and 11 of Sulusaray and Artova District have Cr accumulation group. On the other hand, the other micro catchments codded as 9, 15, 20, 21and located around Zile, Reşadiye and Erbağ Districts were detected less affected or no contaminated by heavy metal (Figure 4). Moreover, Cu have (106.11 mg kg^{-1}) over threshold level in each one soil sample in micro catchments coded as 8 and 11. This case can be assessed no potential risk in terms of heavy metal concentration. Because, these heavy metals don't show grouping of samples as Ni or Cr in Figure 4.

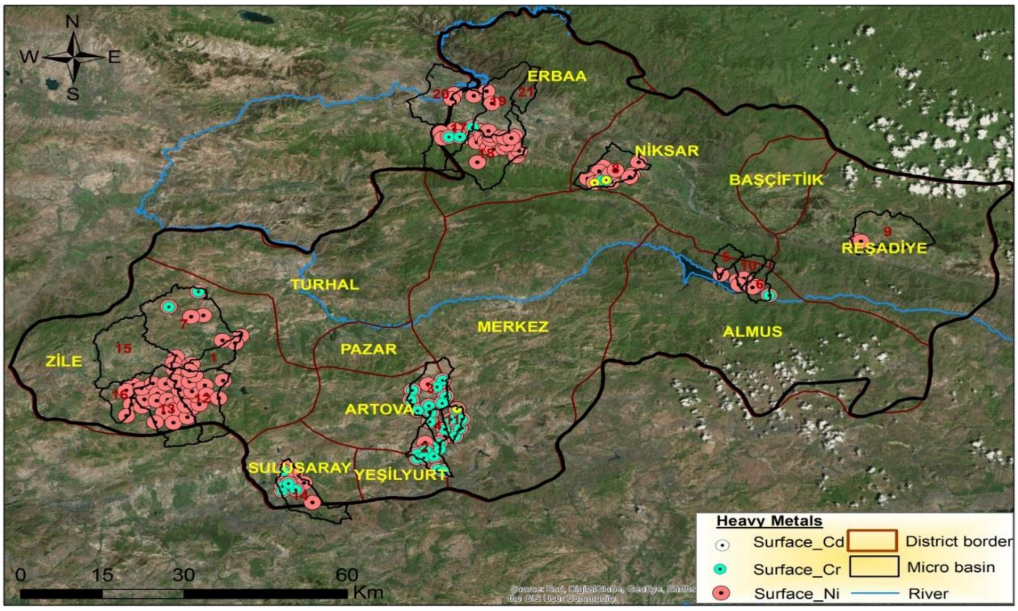

Figure 4. Exceeded heavy metals their maximum permissible concentration in representative micro catchments

A correlation analysis was done for the determination of the relationships between the physico-chemical properties of the soils and heavy metals and given in Table 3. Puschenreiter and Horak (2000) stated that the basic soil characteristics, such as the pH and texture, are of great importance in the availability of heavy metals in the soils. It was found that a significant negative correlation was revealed between the content of sand and the Pb concentrations, whereas a positive correlation was determined between the clay content and the heavy metal content such as Cd and Pb. This result confirmed the data of Temmerman et al. (2003). In addition, no significant relation was found between the content of silt and the heavy metals which are Cd, Pb, Co and Zn. The soil's pH and the $CaCO_3$ content have significant role related to the heavy metals accumulation in the soils. Negative linear correlations were found between these soil properties and some heavy metals in surface soil depth.

Table 3. Relationships between some physico-chemical properties of the soils and the heavy metals in the surface and subsurface soils

Parameters	Sand, %	Clay, %	Silt, %	pH	EC, dS m^{-1}	$CaCO_3$, %	OM, %
Clay, %	-0.740**	1					
Silt, %	-0.506**	-0.206**	1				
pH	-0.019	0.045	-0.030	1			
EC, dS m^{-1}	-0.173**	-0.026	0.285**	0.137*	1		
$CaCO_3$, %	-0.275**	0.090	0.284**	0.374**	0.170**	1	
OM, %	-0.040	0.035	0.014	-0.369**	0.246**	-0.031	1
Cu, mg kg^{-1}	0.073	0.076	-0.204**	-0.155**	-0.241**	-0.432**	-0.024
Cd, mg kg^{-1}	-0.076	0.155**	-0.088	-0.015	0.045	0.084	0.100
Cr, mg kg^{-1}	0.074	0.014	-0.125*	0.079	-0.095	-0.151*	-0.041
Pb, mg kg^{-1}	-0.181**	0.187**	0.023	-0.002	0.048	0.006	0.183**
Co, mg kg^{-1}	0.106	-0.116	-0.006	0.085	-0.092	-0.129*	-0.038
Ni, mg kg^{-1}	0.074	0.034	-0.151*	0.099	-0.075	-0.130*	-0.058
Zn, mg kg^{-1}	-0.008	0.064	-0.070	-0.054	-0.070	-0.216**	0.112

*$P < 0.05$, ** $P < 0.01$, EC: Electric conductivity, OM: Organic matter.

In addition to obtain real heavy metals' values, all the elements were additionally grouped into five levels by Sutherland (2000) in order to estimate their relative accumulation according to the enrichment factors (EF) values for surface and subsurface soil samples. Some statistical characteristics of the EF for the surface and subsurface soils are given in Table 4. In surface soil samples, the EF values for Cd and Ni attest that the soils were enriched with these elements as compared to their mean background value. , Cd, Ni and Cr have found 16%, 10% and 6% soil samples as moderate enrichment class, whereas 55% and 1% of the total soil samples were determined significant enrichment class in terms of Cd and Ni elements. The EF values for the other elements were < 2. The maximal EF value for Cd (21.6) points to the enrichment of the soil with this element; the EF values <2 for Pb, Co, and Zn pointed to the absence of the soil's enrichment with these elements (Table 4). Only a few soil samples have moderate EF classes in terms of Pb and Cu.

Table 4. Some statistical characteristics of the enrichment factors (EF) for the surface and subsurface soils, mg kg^{-1}

Heavy Metals	Mean	SD	*CV	Variance	Min.	Max.	**Skewness	Kurtosis
Cu	0.73	0.31	42.79	0.10	0.14	2.12	1.14	2.95
Cd	0,49	0,26	53,44	0,07	0,00	21.6	0,36	-0,05
Cr	0.59	0.51	86.53	0.26	0.00	3.77	2.61	9.68
Pb	0.53	0.58	110.25	0.34	0.00	9.12	11.79	174.47
Co	0.43	0.28	64.71	0.08	0.00	1.38	0.80	0.61
Ni	0.84	0.85	101.22	0.72	0.03	6.52	3.30	14.11
Zn	0.48	0.16	32.79	0.02	0.05	1.02	0.18	0.18

Conclusion

This present study was conducted in some intensive cultivated land of Tokat Province in order to determine heavy metals content and their enrichment factors. For this purpose, 280 soil samples were collected form surface soil. The results showed that mostly the concentration of Ni followed by Cr exceeded their threshold levels. The local pollutions from Ni and Cr were attributed to the natural influences (particularly due to parent material). The concentrations of the other HMs are relatively lower than the critical values. The mean values of the HMs contents arranged in the following decreasing order: Ni>Cr>Cu>Zn>Co>Pb>Cd in the studied soil sample. In addition, it was found significantly positive relation between Pb and OM while the same relation was also found clay content and Cd and Pb. On the other hand, according to EF of HMs in total soil samples, Cd, Ni and Cr have found 16%, 10% and 6% soil samples as moderate enrichment class, whereas 55% and 1% of the total soil samples were determined significant enrichment class in terms of Cd and Ni elements. Besides, all other HM elements did not exceed minimal enrichment level. However, in some regions of the study area, the Cu, Cd and Pb contents were also slightly raised, this case possibly stems from anthropogenic effects such as excessive P fertilization, field traffic and pesticide using.

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