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Effects of Land Use on the Concentrations of Some Heavy Metals in Soils of Golestan Province, Iran

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ABSTRACT

Soil contamination significantly reduces environmental quality and affects human health. To investigate and assess the effects of land use on the concentrations of some heavy metals in surface soils of Golestan province, 227 soil samples (0-60 cm) were collected from three types of land uses including agricultural lands, natural, and industrial areas. The total metals and metalloids (Cr, Se, As, Cd, Ni, Pb, Zn, Cu, Fe) were extracted and their concentrations were measured in all samples. The results showed that heavy metals accumulations in soil samples of the industrial land uses were higher than agricultural and natural land uses. There was significant correlation among the soils heavy metals (more than 30% for most samples) and also between soil heavy metals and organic carbon content in different types of land uses (average of 40%). Cluster analysis revealed that As and Se had the highest concentration values compared to their corresponding background in most samples and showed the evidence of anthropic effects. Various indices including pollution load index (PLI), modified contamination degree (mC_d), and enrichment factor (EF) were used to determine the soil contamination level. The results of PLI and mC_d, indicated the higher accumulation of heavy metals at industrial land uses. The enrichment factor of Se and As in soils were higher than the other metals, however, their values showed that anthropogenic activities had not serious effects on the environment quality in the studied area.

Keywords: Enrichment factor, Modified contamination degree, Pollution load index.

INTRODUCTION

Land uses have been defined as "the arrangements, activities and inputs people undertake in a certain land cover type to produce, change or maintain it". Land uses and land management practices have a major impact on natural resources including water, soil, nutrients, and plants. Land uses information can be used to develop solutions for natural resource management issues such as salinity and water quality. Deforestation (Khormali *et al*., 2009), urban development, agriculture, and other human activities have substantially altered the earth's landscape (Ayoubi *et al.,* 2011; Khormali and Nabiollahi, 2009). Such disturbance on the land, affect the important ecosystem processes and services, which can have wide–ranging and long–term consequences.

Soil pollution, especially with heavy metals, due to changes in the land uses pattern over the last few decades, has become an important environmental issue in developed countries (Adriano, 2001). Soil contamination by heavy metals in agricultural land uses is associated with the use of fertilizers, pesticides, and herbicides by farmers to increase crop yields. These also contaminate the land when they are washed up into the soil. Although some trace elements are essential for plant nutrition, plants growing in the close vicinity of industrial areas display increased concentration of heavy metals, serving in many cases as biomonitors of pollution loads (Mingorance *et al*., 2007; Yang *et al*., 2005; Dankoub *et al*., 2012).

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The use of agrochemicals such as pesticides and fertilizers may have resulted in undesirable accumulation of trace elements, such as arsenic, cadmium, copper, lead, and zinc in soils (Merwin *et al*., 1994; Van Gaans *et al*., 1995; Harris *et al*., 2000). Numerous studies have already demonstrated that areas close to industrial activities are marked by noticeable contamination of air, soil and water (Landajo *et al*., 2004; Kocher *et al*., 2005; Karimi *et al*., 2011; Naimi and Ayoubi, 2013). Hence, such activities can affect the air we breathe, the water we use, and the soil we stand on and can ultimately lead to illness and/or harm to the residents in the affected area. There is an increasing awareness that heavy metals present in soil may have negative effects on human health and on the environment (Abrahams, 2002; Mielke *et al*., 2005; Selinus *et al*., 2005; Uba *et al*., 2009; Dauvalter *et al*., 2009; Venugopal *et al*., 2009). One of the most crucial properties of metals is that they are not biodegradable in the environment (Harte *et al*., 1991; Schuurmann and Market, 1998; Sundararajan and Natesan, 2010; Taghinia Hejabi *et al*., 2010).

Assessing soil pollution with toxic elements has to be done compared to the baseline concentrations in soil. Pollution, in this case, will be measured as the amount of metal enrichment in the sample, above the concentrations present in the background value (Abrahim and Parker, 2008; Rafiei *et al*., 2010; Razos and Christides, 2010). In order to assess the impact of toxic metals pollution on different environments by using various enrichment calculation methods, several works have been done (Abrahim and Parker, 2008; Adomako *et al*., 2008; Aikpokpodion *et al*., 2010; Ghrefat *et al*., 2010, Liu *et al*., 2005; Nasrabadi *et al*., 2010). Some indicators of contamination in soil and sediment most often applied in these studies are enrichment factor (EF), pollution load index (PLI), modified contamination degree (mC_d) and geoaccumulation index. This approach would help adopt an effective effluent management strategy towards control over enhanced metal levels with recycling of effluents for toxic metal separation and soil remediation and reclamation. The objectives of the present study were (1) to determine contents of Cr, Se, As, Cd, Ni, Pb, Zn, Cu and Fe in soils of different land uses; (2) to determine soil contamination indices and assess the soil pollution, and (3) to determine their natural or anthropogenic sources, using multivariate analysis.

MATERIALS AND METHODS

Study Area Description

The study was carried out in Golestan province, located in north east of Iran, south of Caspian sea with an area of about 20,893 km^2 (Figure 1). The climate of the province is variable with the average annual temperature of 18.2^{0}C and the annual rainfall ranges from 250 to 700 mm. The soil moisture and temperature regimes are Aridic and Thermic, respectively. Based on soil taxonomy (USDA Soil Taxonomy, 2010), most soils are classified as Mollisols and Aridisols.

Wheat, cotton, and summer crops are the main products in Golestan and the area is one of the

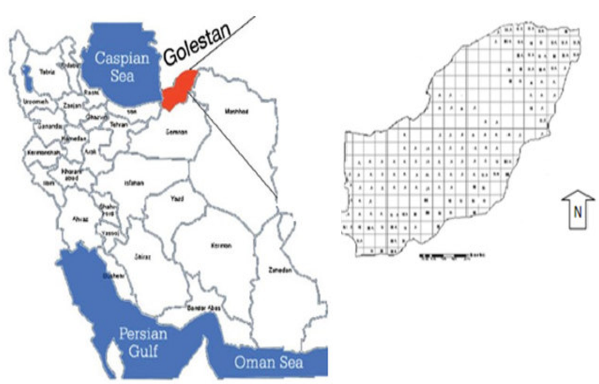

Figure 1. Location of the study area in Golestan province, northern Iran.

most important parts of the country due to extensive agricultural activities. Industries are also short aged and soils are mostly fertile in central parts of the region. This study focused on arable lands as agricultural land in central parts of the province where agricultural activities are intensive. Other types of farming in other parts (north and south) are local and sparse and have no considerable effect on regional soil pollution. The soil quality in central parts is influenced by geological materials from mountainous regions in the south and vast aeolian dry lands (loess deposits) in the north. The main lithologic units in southern regions are igneous and metamorphic rocks while northern parts are composed of vast thick loess deposits.

The study included four adjacent land parcels under different uses: (1) Natural lands containing pasture and undisturbed soils covered by Artemisia, Salicornia and Astragallus species; (2) Agricultural lands containing cultivated and fallow lands with different products, and (3) Industrial lands containing the soils next to the factories and mines. The study area was divided into regular lattices with 10 km^2 and soil samples were collected from each map lattice (Figure 1).

Data Collection and Soil Analysis

After preliminary studies of the topographic maps (1:25,000), the study location was delineated using GPS. Two hundred and twenty seven soil samples were collected from 0-60 cm depth of the study area. The samples were collected from 117, 60, and 50 points for agricultural, natural and industrial lands, respectively, covering almost the whole provincial area (Figure 2).

Soil samples were air-dried and passed through a standard 2 mm sieve. Soil chemical properties such as electrical conductivity (EC), pH, organic carbon percentage, cation exchange capacity (CEC), and calcium carbonate percentage were determined using standard frequently used methods (Sparks *et al*., 1996). Then, samples were digested with HNO_3 and H_2O_2 using Method 3050B (USEPA, 1996). Concentrations of some metals including Cr, Se, As, Cd, Ni, Pb, Zn, Cu and Fe were determined in the digested solutions, using inductively coupled plasma-atomic emission spectroscopy (ICP-AES). The minimum detection limit (MDL), which is defined as the minimum concentration of substance that can be measured and reported with 99% confidence, was determined using EPA 40CFR Part 136, Appendix B. Standard reference materials soils were obtained before the surveys (Cabrera *et al*., 1999; Liu *et al*., 2005) and they were verified with National Institute of Standards and Technology (NIST) traceable certified reference standards.

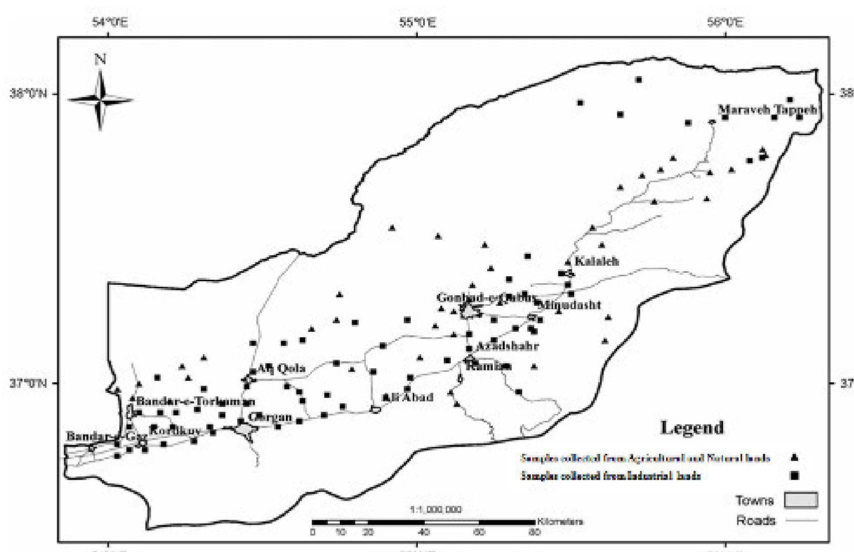

Figure 2. Geographical distribution of sampling points.

Statistical Analysis

The distribution of data was examined by the Kolmogorov-Smirnov test. Statistical evaluation and mean comparisons were performed by analysis of variance (ANOVA), LSD test, and Pearson's rank correlation (data with normal distribution) at 0.05 significance level using SPSS 19 statistical software. To evaluate the analytical data, correlation analysis and cluster analysis (CA) were also used. The Pearson correlation coefficient (r) was used to measure the strength of a linear relationship between different metals. Cluster analysis was used to elucidate the latent relationships between heavy metals in soils under the same kind of land uses in the studied area. Hierarchical cluster analysis was also performed using the following settings: the linkage type used was nearest neighbor and the distance method was the Pearson correlation.

Cluster analysis was used to exhibit element concentration in different lands (Luo *et al*., 2007) and identify the source and relatively homogeneous groups of heavy metals (Hu *et al*., 2013; Franco Uria *et al*., 2009).

Evaluation Method

Based on the results obtained, the Tomlinson pollution load index (PLI) was computed for surface soil samples to detect the relative heavy metal accumulation and environmental quality of the soils in the study area. The *PLI* index (Angulo, 1996) is achieved by

$$\mathrm{PLI} = \sqrt[n]{\mathrm{CF}_1 \times \mathrm{CF}_2 \times \ldots \times \mathrm{CF}_n} \quad (1)$$

Where, *CF* (Contamination Factor) is the ratio of the concentration of each metal (C_{metal}) to the background concentration ($C_{background}$) in the soil as:

$CF = C_{metal}/C_{background}$ (2)

In this relationship, CF_1 to CF_n indicates the contamination factors calculated for the first soil sample to the nth one. A *PLI* value close to one, indicates heavy metal loads near the background level, while values above one, indicate soil pollution (Cabrera *et al*., 1999; Liu *et al*., 2005).Therefore, soils with *PLI* value of more than 1 are polluted, whereas values less than 1 indicate no pollution (Harikumar *et al*., 2009). A modified and generalized form of Hakanson (1980) equation for the calculation of the overall degree of contamination was defined as the sum of all contamination factors (CF) for a given set of pollutants divided by the number of analyzed pollutants as follows (Abrahim, 2005):

$$\mathrm{mC_d} = \frac{\sum \mathrm{CF}}{n} \quad (3)$$

Where, *CF* is contamination factor and n is the number of analyzed elements. For the descriptive classification of the modified contamination degree (mC_d), the following gradations are proposed (Table 1).

The enrichment factor (EF) is the relative abundance of a chemical element in a soil compared to the reference matter. *EFs* are calculated based on different reference materials such as earth crust (Krishna and Govil, 2005). The enrichment factor is calculated by comparing the concentration of a tested element with that of a reference element (Liu *et al*., 2005). The value of the enrichment factor was calculated using the following modified relationship based on the equation suggested by Buat- Menard and Chesselet (1979):

$$\mathrm{EF} = \frac{\frac{C_n(\mathrm{sample})}{C_{ref}(\mathrm{sample})}}{\frac{B_n(\mathrm{background})}{B_{ref}(\mathrm{background})}} \quad (4)$$

Where, C_n (sample) is the content of the examined element in the examined environment, C_{ref} (sample) is the content of the

Table 1. Different modified degree of contamination (mC_d) for soil (Abrahim and Parker, 2008).

mC_d Class	Modified Degree of Contamination Level
$mC_d < 1.5$	Nil to very low degree of contamination
$mC_d < 2 \leq 1.5$	Low degree of contamination
$mC_d < 4 \leq 2$	Moderate degree of contamination
$mC_d < 8 \leq 4$	High degree of contamination
$mC_d < 16 \leq 8$	Very high degree of contamination
$mC_d < 32 \leq 16$	Extremely high degree of contamination
$mC_d \geq 32$	Ultra high degree of contamination

reference element in the examined environment, B_n (background) is the content of the examined element in the reference environment, and B_{ref} (background) is the content of the reference element in the reference environment.

The reference element is assumed to have little variability of occurrence and is present in trace concentration in the examined environment. It is also possible to use a geochemically characteristic element which is present in the environment in high concentration, but is characterized by none of these effects i.e. synergism or antagonism towards the examined element (Loska *et al*., 2004). The most common reference elements are Sc, Mn, Ti, Al, and Fe (Buat- Menard and Chesselet, 1979; Martin and Meybeck, 1979; Li, 1981; Reinmann *et al*., 2000; Schiff and Weisberg, 1999; Sutherland, 2000; Tomza *et al*., 1982; Pacyna and Winchester, 1990; Quevauviller *et al*., 1989). In the present study, Al ratio was regarded orderly as reference element and reference environment. Five contamination categories were recognized on the basis of the enrichment factor as follows (Table 2).

RESULTS AND DISCUSSION

Effects of Land Uses on the Concentrations of Metals and Soil Characteristics

Descriptive statistical data including mean, standard deviation, minimum and maximum of some measured soil parameters such as heavy metals concentrations (Cr, Se, As, Cd, Ni, Pb, Zn, Cu and Fe) and some chemical characteristics (EC, pH, OC, CEC and $CaCO_3$) were determined (Table 3). The description of soil chemical properties showed that soil electrical conductivity had low values. only salinity in agricultural lands showed significant increase compared to the other land uses. It could be due to agricultural management and irrigation with saline water. Acidity values did not show significant changes in different land uses. Also, its variation was very low which suggested that heavy metals accumulation did not have any effects on soil pH. On the other hand, pH values could not vary appreciably due to high lime content. Organic carbon percentage was low in different types of land uses without significant changes (Liu *et al*., 2003). Cation exchange capacity showed similar values in different land uses. Lime percentage showed high values, especially in natural lands that had lithogenic origin (Table 3).

In most samples, agricultural lands showed the moderate amounts of Cr, Se, As, Cd, Ni, Pb, Zn, Cu and Fe compared to industrial and natural lands which indicated the highest and lowest values in most samples. The mean values of heavy metals contents in all types of land uses were lower than the world median value (Bowen, 1979).

The results of one-way analysis of variance for heavy metals and soil chemical properties in different land uses showed that there were significant differences between heavy metals and soil characteristics, in the three types of land uses, except for pH and OC content. The results of comparing heavy metals also showed high concentrations in industrial lands, compared to agricultural as well as natural lands, respectively. Arsenic and Se showed the highest values of concentrations in agricultural lands and suggested that there were several locations having great As and Se concentrations, revealing that soils in some areas were more contaminated. The

Table 2. Different modified degrees of Enrichment Factor (EF) for soil (Kargar *et al*., 2012)**.**

EF class	Modified Degree of Enrichment Factor
EF< 2	Deficiency to mineral
EF< 5 ≤ 2	Moderate enrichment
EF< 20 ≤ 5	Significant enrichment
EF< 40 ≤ 20	Very high enrichment
EF ≥ 40	Extremely high enrichment

Table 3. The concentrations of heavy metals and soil chemical properties in soils from different land uses.[a]

Soil characteristics	Land uses	Mean	SD	Minimum	Maximum
Cr (mg kg^{-1})	Agricultural land (n=117)	60.68[a]	10.20	38	83
	Uncultivated land (n=60)	53.13[b]	14.57	23	91
	Industrial land (n=50)	60.82[b]	15.21	28	88
Se (mg kg^{-1})	Agricultural land (n=117)	0.56[a]	0.21	0.17	1.06
	Uncultivated land (n=60)	0.36[b]	0.07	0.25	0.58
	Industrial land (n=50)	0.37[b]	0.11	0.21	0.64
As (mg kg^{-1})	Agricultural land (n=117)	9.53[a]	2.01	3.5	15.57
	Uncultivated land (n=60)	8.15[b]	2.46	2.10	14.42
	Industrial land (n=50)	9.41[a]	2.16	3.44	12.87
Cd (mg kg^{-1})	Agricultural land (n=117)	0.18[ab]	0.05	0.12	0.33
	Uncultivated land (n=60)	0.16[b]	0.07	0.04	0.35
	Industrial land (n=50)	0.19[a]	0.06	0.07	0.36
Ni (mg kg^{-1})	Agricultural land (n=117)	36.24[b]	7.10	20.40	53.20
	Uncultivated land (n=60)	38.67[b]	9.51	19.50	56.70
	Industrial land (n=50)	46.88[a]	10.92	21.70	73.70
Pb (mg kg^{-1})	Agricultural land (n=117)	13.05[c]	3.15	7.30	21.80
	Uncultivated land (n=60)	17.04[b]	5.84	6.90	31.30
	Industrial land (n=50)	19.42[a]	7.48	8.50	44
Zn (mg kg^{-1})	Agricultural land (n=117)	70.06[c]	15.63	41.90	111.50
	Uncultivated land (n=60)	80.47[b]	22.97	25.10	127.20
	Industrial land (n=50)	91.17[a]	16.82	48.80	114.2
Cu(mg kg^{-1})	Agricultural land (n=117)	22.99[b]	5.28	11.50	35.90
	Uncultivated land (n=60)	22.35[b]	6.63	11.30	38.70
	Industrial land (n=50)	27.76[a]	8.45	11.9	45.6
Fe (mg kg^{-1})	Agricultural land (n=117)	33017.82[b]	5149.34	20767	44214
	Uncultivated land (n=60)	32575.85[b]	7849.01	21397	51450
	Industrial land (n=50)	38688.42[a]	10071.59	19947	63167
EC (ds m^{-1})	Agricultural land (n=117)	3.77[b]	4.65	0.36	17.70
	Uncultivated land (n=60)	1.88[b]	2.32	0.34	11.7
	Industrial land (n=50)	8.55[a]	8.55	0.45	95.2
pH	Agricultural land (n=117)	7.94[a]	0.21	7.05	8.49
	Uncultivated land (n=60)	8[a]	0.28	7.08	8.9
	Industrial land (n=50)	8.01[a]	8.01	7.24	8.53
OC (%)	Agricultural land (n=117)	1.07[a]	0.41	0.35	2.04
	Uncultivated land (n=60)	1.19[a]	0.78	0.13	3.08
	Industrial land (n=50)	1.11[a]	1.11	0.20	2.85
CEC (meq 100 g^{-1})	Agricultural land (n=117)	21.79[a]	9.12	6.12	46.08
	Uncultivated land (n=60)	19.47[a]	12.23	4.60	60
	Industrial land (n=50)	16.84[a]	14.84	5.91	33.04
$CaCO_3$ (%)	Agricultural land (n=117)	16.85[b]	7.15	1.76	36.4
	Uncultivated land (n=60)	20.03[a]	11.41	3.75	41.5
	Industrial land (n=50)	12.92[c]	12.92	1.50	26.5

[a] Land uses followed by different letters are significantly different at the $P< 0.05$ level.

concentrations of Zn and Pb in natural and industrial lands showed higher values than agricultural lands. The ranges of Se and Cd were also lower than the other metals (Table 4).

Correlation analysis based on the Pearson coefficient was conducted to determine the extent of the relationships among metals in soils of different land uses. The correlation between soil heavy metals (Table 5) and soil

Table 4. ANOVA of heavy metals and soil chemical properties for different land uses.

Soil characteristics	F	Significance
Cr	2.95	0.06
Se	6.74	0.00
As	9.97	0.00
Cd	18.80	0.00
Ni	0.43	0.66
Pb	3.76	0.03
Zn	20.92	0.00
Cu	1.86	0.16
Fe	8.80	0.00
EC	4.37	0.02
pH	9.18	0.00
OC	2.81	0.07
CEC	5.84	0.003
$CaCO_3$	0.17	0.86

Table 5. Correlations between heavy metals contents in different land uses.

		Cr	Se	As	Cd	Ni	Pb	Zn	Cu	Fe
	Agricultural land	1								
Cr	Natural land	1								
	Industrial land	1								
	Agricultural lLand	0.38**	1							
Se	Natural land	0.15	1							
	Industrial land	0.14	1							
	Agricultural land	0.46**	0.14	1						
As	Natural land	0.44**	0.12	1						
	Industrial land	0.28**	0.02	1						
	Agricultural land	0.32**	0.08	0.28**	1					
Cd	Natural land	0.22*	0.21*	0.33**	1					
	Industrial land	0.09	0.17	0.20*	1					
	Agricultural land	0.89**	0.35**	0.47**	0.36**	1				
Ni	Natural land	0.74**	-0.02	0.39**	0.39**	1				
	Industrial land	0.75**	-0.13	0.23*	0.19*	1				
	Agricultural land	0.68**	0.28**	0.40**	0.34**	0.66**	1			
Pb	Natural land	0.36**	-0.09	0.21*	0.27**	0.65**	1			
	Industrial land	0.24**	-0.21*	0.30**	0.23*	0.55**	1			
	Agricultural land	0.75**	0.50**	0.29**	0.35**	0.82**	0.71**	1		
Zn	Natural land	0.36**	0.12	0.03	0.22*	0.58**	0.61**	1		
	Industrial land	0.43**	-0.02	0.17	0.34**	0.72**	0.67**	1		
	Agricultural land	0.84**	0.37**	0.38**	0.29**	0.91**	0.70**	0.82**	1	
Cu	Natural land	0.67**	0.06	0.27**	0.27**	0.77**	0.37**	0.40**	1	
	Industrial land	0.76**	-0.09	0.17	0.21*	0.82**	0.46**	0.64**	1	
	Agricultural land	0.80**	0.40**	0.55**	0.31**	0.79**	0.70**	0.79**	0.70**	1
Fe	Natural land	0.59**	0.09	0.26**	0.08	0.58**	0.39**	0.47**	0.57**	1
	Industrial land	0.62**	-0.07	0.21*	0.10	0.70**	0.40**	0.58**	0.67**	1

* Correlation is significant at the 0.05 level (2-tailed), ** Correlation is significant at the 0.01 level (2-tailed).

chemical characteristics (Table 6) in agricultural lands (P< 0.01) were higher than their values in natural and industrial lands. The results showed that Se correlation with other metals in different land uses was the lowest, suggesting perhaps different soil sources for Se. Other metals such as Cr, Ni, Pb and Cu were closely related to each other, which could return to similar origin in different land uses (Table 5).

Table 6. Correlations between heavy metals contents and soil chemical characteristics in different land uses.

		EC	pH	OC	CEC	$CaCO_3$
	Agricultural land	-0.17	-0.24*	0.52**	0.10	-0.06
Cr	Natural land	-0.07	-0.12	0.32**	0.35**	-0.42**
	Industrial land	-0.13	-0.13	0.42**	0.21*	-0.32**
	Agricultural land	0.11	0.07	0.59**	0.04	0.23*
Se	Natural land	0.06	0.17	0.24*	-0.02	0.18
	Industrial land	-0.07	0.06	0.31**	0.25**	0.38**
	Agricultural land	-0.09	-0.01	0.33**	-0.14	0.07
As	Natural land	0.03	-0.18	0.30**	0.04	-0.08
	Industrial land	-0.06	-0.04	0.14**	0.02	-0.05
	Agricultural land	-0.31**	-0.09	0.20*	0.12	-0.03
Cd	Natural land	-0.22*	0.14	0.38**	0.07	0.13
	Industrial land	-0.05	0.04	0.37**	0.11	0.16
	Agricultural land	-0.20*	-0.17	0.48**	0.10	-0.08
Ni	Natural land	-0.27**	-0.06	0.48**	0.28**	-0.24*
	Industrial land	-0.11	0.04	0.43**	-0.07	-0.37**
	Agricultural land	-0.30**	-0.23*	0.49**	0.32**	-0.17
Pb	Natural land	-0.24*	-0.02	0.50**	0.12	-0.06
	Industrial land	-0.10	-0.02	0.34**	-0.20*	-0.33**
	Agricultural land	-0.21*	-0.06	0.55**	0.06	0.05
Zn	Natural land	-0.13	0.16	0.36**	0.07	-0.06
	Industrial land	-0.09	0.15	0.35**	-0.16	-0.29**
	Agricultural land	-0.17	-0.28**	0.50**	0.14	0.12
Cu	Natural land	-0.14	-0.14	0.31**	0.22*	-0.29**
	Industrial land	-0.06	-0.02	0.38**	-0.06	-0.39**
	Agricultural land	-0.23*	-0.08	0.57**	0.09	0.03
Fe	Natural land	-0.20	-0.08	0.39**	0.22*	-0.19*
	Industrial land	-0.18	0.20*	0.14	-0.12	-0.40**

* Correlation is significant at the 0.05 level (2-tailed), ** Correlation is significant at the 0.01 level (2-tailed).

Multivariate Analysis

The simple correlations between soil chemical characteristics and soil heavy metals concentrations in different land uses are shown in Table 6. The results showed that compared to other soil characteristics, organic carbon percentage had the highest correlation and pH had the lowest correlations with soil heavy metals (Dayani and Mohammadi, 2010). The correlation values between different metals with organic carbon were similar and there were no significant correlation difference between several heavy metals with organic carbon. Soil organic carbon increment can lead to elevated soil adsorption capacity by which accumulation of heavy metals would be enhanced (Gao *et al*., 1997). The correlation analysis provides little information about the sources of metals. Therefore, cluster analysis was also performed on heavy metals concentrations in agricultural, natural and industrial lands, using the nearest neighbor linkage method based on correlation coefficients. The results are illustrated as dendrograms in Figures 3(a-c). The distance cluster represents the degree of association between elements. The smaller value on the distance cluster indicated higher significance in association. A criterion for distance clusters requiring between 10 and 15 was adopted.

In agricultural lands, two distinct clusters were identified (Figure 3-a). Cluster 1 contained Ni, Se, Fe, As, Cr, and Cd. The concentrations of Ni, Fe, Cr and Cd in soils of agricultural

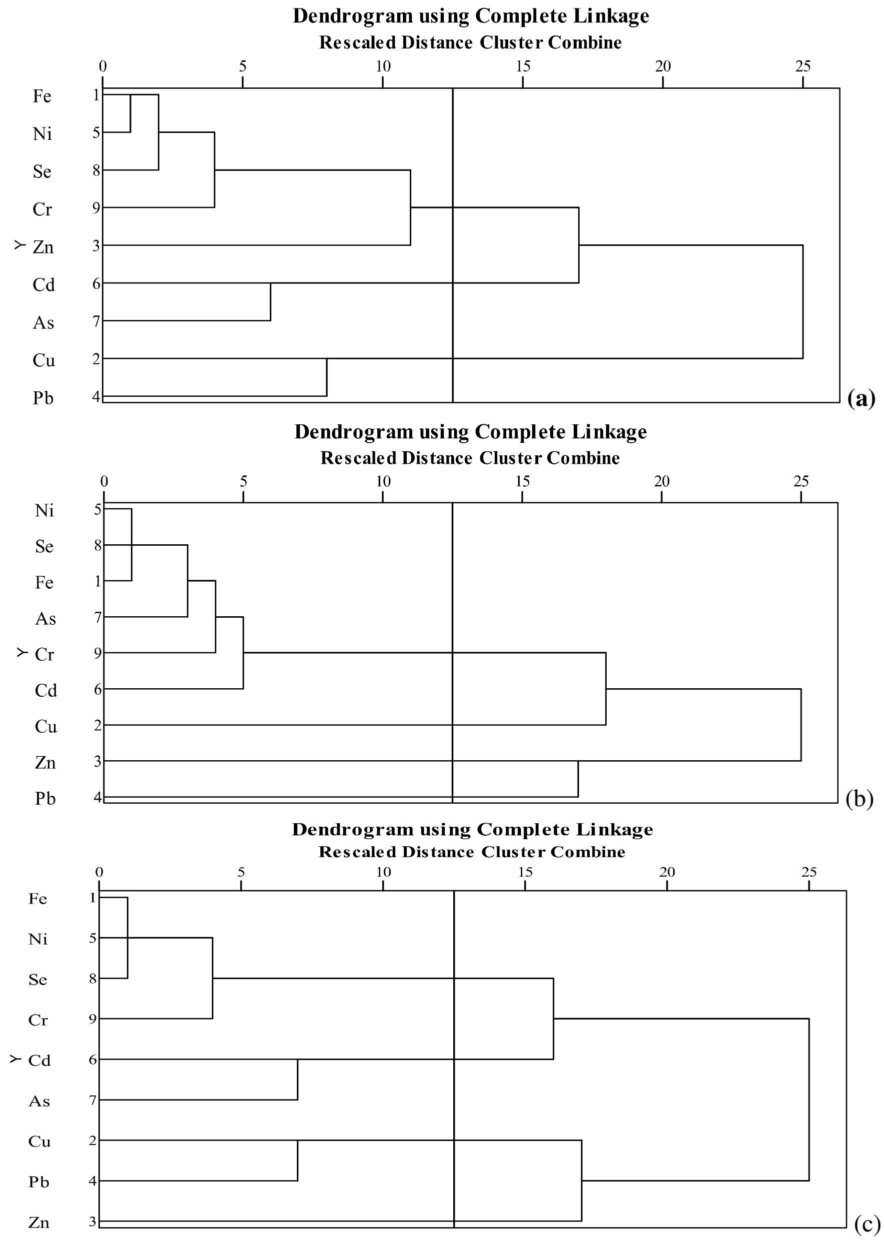

Figure 3. Dendrogram of the cluster analysis of (a) agricultural lands (b) natural lands (c) industrial lands based on heavy metals concentrations.

lands were less than their corresponding background concentrations, it could be concluded that they originated from parent materials, and anthropogenic activities had no important effects on them. Moreover, the mean concentrations of As and Se were high in different types of land uses, especially As that showed the highest concentrations. This could be due to excessive use of phosphate fertilizers that contained As and also widespread applications of pesticides as well as fungicides containing Se in the studied area (Yang *et al.*, 2005; Ma and Liu, 1998; Wang, 2000; Taylor, 1997).

In addition, irrigation of agricultural lands by wastewaters and also sludge applications are likely important sources of Se in agricultural lands (Environment Agency, 2009). In summary, these metals probably came from natural soil parent materials, application of pesticides and fertilizers, as well as wastewater irrigation.

Cluster 2 of agricultural lands contained Cu, Zn and Pb which originated from the natural soil parent materials due to showing lower values than the limited range of its background concentrations. Holland and Solomona (1999) proposed that elevated soil copper concentrations in orchard soils arise from the long-term use of copper-based fungicides. Comparison of mean concentrations for Cu, Zn and Pb, suggested that industrial lands had higher concentrations of heavy metals than agricultural and natural lands, which could be due to the industrial activities releasing smoke, wastewater, and other similar effluents (Figure 3-a). The results revealed that Cu, Zn, Pb, Cd and AS accumulated in agricultural fields whereas Ni, Cr, Co and Fe were controlled by soil parent material compounds (Kelepertzis, 2014).

In natural lands, three distinct clusters were identified (Figure 3-b). Cluster 1 contained Fe, Ni, Se, Cr and Zn. The concentrations of these metals were found in most samples in different land uses to be the lowest, except Zn that showed the average concentration. It should be emphasized that the concentrations of these metals in natural lands still represent a considerable absolute quantity that may indicate the role of parent materials for Fe, Ni, Se and Cr accumulations.

Cluster 2 of natural lands contained Cd and As. The concentrations of As and Cd were lower in different land use patterns. Cadmium values were less than their corresponding background concentrations and this can prove the role of lithogenic origin and parent materials. In contrast, As concentration showed higher values than their corresponding background concentrations and suggested the effects of human activities. Cluster 3 contained Cu and Pb exhibited lower concentrations than their corresponding background while Pb concentration showed higher values than agricultural lands that can be related to the dumping of solid wastes and domestic rubbish (Figure 3-b).

In industrial lands, three distinct clusters were identified (Figure 3-c). Cluster 1 contained Fe, Ni, Se and Cr. Except selenium, which in industrial lands showed lower values than agricultural lands, other metals had the highest concentrations in the industrial lands in most samples. Selenium also showed higher accumulation than its corresponding background concentrations. Cluster 2 contained Cd and As, and showed the highest accumulation of As compared to their reference values and their values in different land uses. Cluster 3 contained Cu, Pb, and Zn with significant increase of its metal concentrations over other land uses, though their accumulations were lower than their reference values. It might be due to ingredients discharged into the soil such as seepage from a landfill, percolation of contaminated water into the soil, rupture of underground storage tanks, solid waste seepage, and other anthropogenic activities. Franco Uria *et al*. (2009) used cluster analysis to identify the source of heavy metals. Their results showed that Mn, Co, Ni, Cu, Fe, and Cr were associated in two lithogenic components, while an anthropogenic origin attributed to slurry applications was identified for Cd, Pb and Zn. They divided heavy metals to geological origin (Ni, Cr, Co, Mn and Fe) and anthropic origin (Pb, Zn and Cu) by multivariate analysis (Kaitantzian *et al*., 2013). Multivariate and geostatistical analysis suggested that soil Cr, Ni and Zn had a lithogenic origin, while Cu and Pb concentrations showed industrial and agronomic origins (Sun *et al*., 2013). To identify the source of eight hazardous heavy metals in agricultural soils, they used multivariate statistical techniques and enrichment factor and showed that Cr, Ni, Cu, Zn, and Pb mainly originated from a natural source while, Cd, As and Zn mainly came from agricultural practices and, finally, Hg and Pb originated from industries and traffic sources (Cai *et al*., 2012).

Ecological Risks Assessment of Metals in Different Land Uses

The assessment of soil contamination was conducted using the pollution load index (PLI), modified contamination degree (mC_d) and enrichment factor (EF). In the version suggested by Hakanson (1980), an assessment of soil contamination was conducted through reference of the concentrations in the surface layer. Based on the *PLI*, the soils of agricultural lands, natural lands and industrial lands were classified as uncontaminated soils with *PLI* values of 0.8, 0.76, and 0.88, respectively. These values indicated higher values of *PLI* in industrial lands than in agricultural and natural lands. The differences between PLI values in different land uses were not significant, though their correlation was high ($r > 0.95$).

The mC_d was computed in different land uses. The results showed that industrial lands with higher mC_d had higher ecological risk while agricultural and natural lands had lower risk. The mC_d for industrial, agricultural and natural lands were 0.92, 0.87, and 0.80, respectively, which showed that all soils in the three different land uses had very low degree of contamination ($mC_d< 1.5$) (Table 1). Survey of the mC_d values in different land uses emphasized that there were no significant differences between the three types of land uses, though their correlations were high ($r > 0.95$).

The assessment of element abundance in a soil compared to the reference matter was based on enrichment factor (EF). Based on the results, the enrichment factors were in the following order: Agricultural lands> Industrial lands> Natural lands. Regarding the values of *EF* in different land uses, results showed that heavy metals in different land uses had low enrichment factor, except As and Se with significant enrichment, especially in agricultural lands (Table 2). Comparing their mean values in different land uses showed that there were no significant differences between them while their correlation were high ($r> 0.90$). Box plot diagrams were used to compare *EF* values between soil samples collected from different land uses (Figures 4a-c). Comparison of the three land uses clearly showed the effects of human activities on the metal concentrations in the surface soils in the studied area. Enrichment factor values greater than one indicated the enrichment corresponding mainly to anthropogenic effects. (Figure 4-a) shows the *EF* values of agricultural lands. The results showed that Se, As and almost Cd values were higher than the other metals such as Cr, Ni, Pb, Zn, Cu, and Fe; and variation of *EF* among them were negligible. The variation of EF in natural lands was higher than agricultural lands. Enrichment factor values of Se and As were high; Cd, Pb and Zn showed moderate values and Cr, Ni, Cu and Fe showed low values (Figure 4-b). This could be related to the role of parent materials. Industrial lands showed the highest *EF* for As and Se, while Cd, Pb, and Zn showed the moderate and Cr, Ni, Cu and Fe showed the lowest values of *EF* (Figure 4-c).

CONCLUSIONS

The present project studied the concentrations of heavy metals in three types of land uses including agricultural, natural and industrial lands of Golestan province. The results indicated that there were no significant heavy metals pollution, except for As and Se and especially in soils of agricultural lands. They were affected by anthropogenic activities. Concentrations of heavy metals in agricultural soils were mostly comparable with natural geochemical background of the studied area. Such pollution probably originates mainly from phosphate fertilizer, sludge applications, as well as pesticides and fungicides uses. Selenium is extensively derived from loess deposits of the area, so natural background in the top layers is the main source of Se. Low contamination of Cr, Cd, Ni, Pb, Zn, Cu, and Fe might have come from soil parent materials. The contamination of the environment was found in the following order of pollution load index and degree of contamination: Industrial lands> Agricultural lands> Natural lands. Although the indexes of soil pollution were similar, but enrichment factor values in agricultural lands were higher than industrial lands. The mean EF values for Se and As in different land uses suggested that

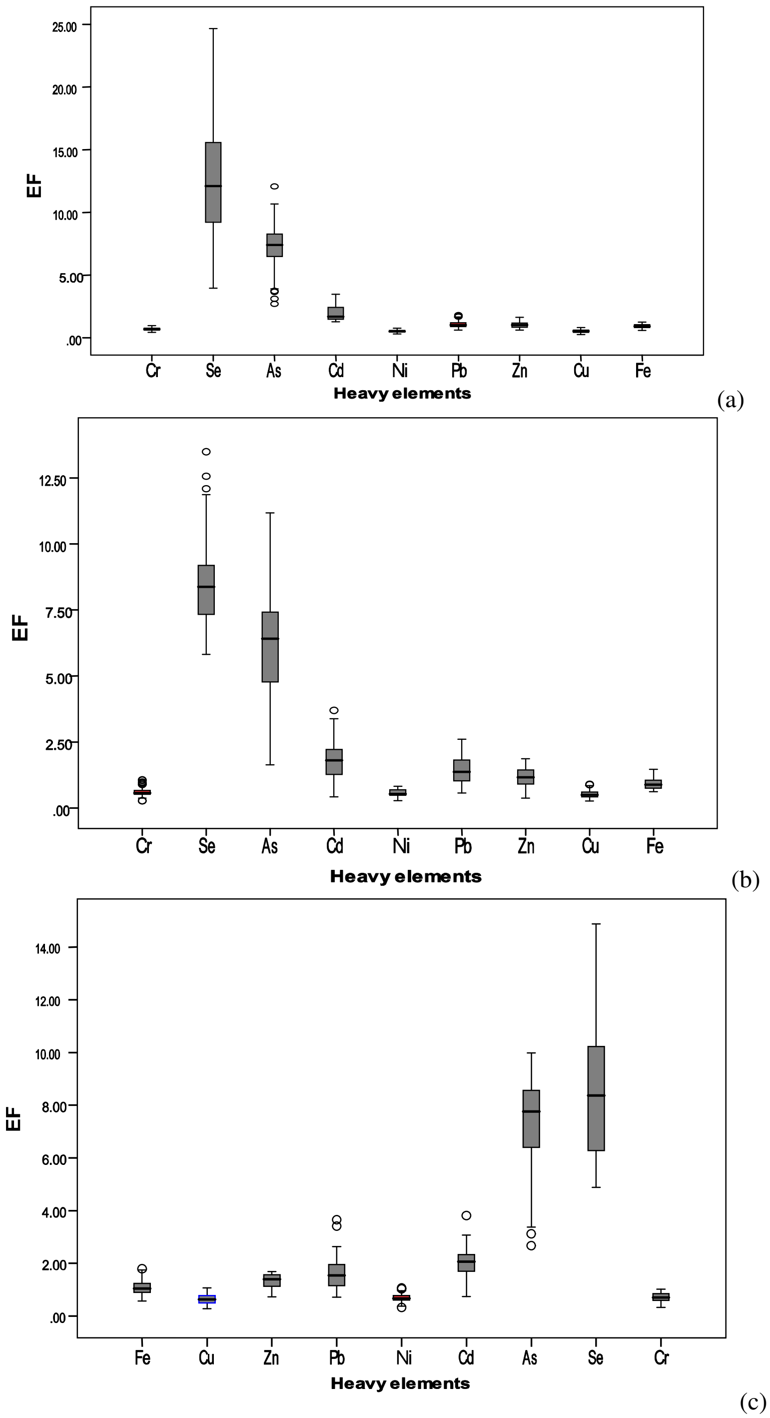

Figure 4. Box plot of enrichment factor (EF) values for heavy metals in (a) agricultural lands (b) natural lands (c) industrial lands.

more attentions need to be given to these metals in order to protect the environment.

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اثرات تغییر کاربری اراضی بر تجمع برخی از فلزات سنگین در خاکهای استان گلستان، ایران

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چکیده

آلودگی خاک به عنوان یکی از عوامل اصلی کاهش کیفیت محیط زیست و تهدیدکننده سلامت انسانها به حساب می آید. برای بررسی اثرات تغییر کاربری اراضی بر تجمع برخی فلزات سنگین در خاکهای سطحی استان گلستان، ۲۲۷ نمونه خاک از عمق ۰–۶۰cm اراضی طبیعی، کشاورزی و صنعتی برداشت شدند. غلظت کل فلزات سنگین شامل (Cr, Se, As, Cd, Ni, Pb, Zn, Cu, Fe) اندازه گیری شدند. نتایج غلظت فلزات سنگین اراضی صنعتی نسبت به اراضی کشاورزی و طبیعی مقادیر بالاتری را نشان می دادند. نتایج ماتریس همبستگی، همبستگی بالا مابین فلزات سنگین خاک (بیشتر از ۳۰ درصد) و همچنین میان فلزات سنگین و کربن آلی خاک (بیش از ۴۰ درصد) در اراضی مختلف را نشان می داد. تجزیه و تحلیل خوشه ای برای گروه بندی فلزات سنگین با توجه به نزدیکی آنها در اراضی مختلف اجرا گردید. در بیشتر نمونه ها، نتایج وجود مقادیر بالای آرسنیک و سلنیوم نسبت به مقادیر مرجع و نقش عوامل انسانی در تجمع آنان را نشان می داد. شاخص های متفاوت نظیر شاخص بار آلودگی (PLI)، درجه آلودگی اصلاح شده (mC_d) و فاکتور غنی شدگی (EF) برای تعیین درجه و سطح آلودگی خاک به کار برده شدند. نتایج به دست آمده از شاخصهای بار آلودگی و درجه آلودگی مقادیر زیادتر آنها را در اراضی صنعتی در مقایسه با سایر کاربری اراضی نشان می داد. همچنین فاکتور غنی شدگی آرسنیک و سلنیوم در خاکهای مورد مطالعه نسبت به سایر فلزات سنگین دارای مقادیر بیشتری بود. به طور کلی مقادیر اندازه گیری شده نشان دهنده نقش ناچیز فعالیتهای انسانی و صنعتی در کاهش کیفیت محیط زیست و آلودگی خاک بود.