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Original Article

Ecological and Human Health Risks Assessment of Potentially Toxic Elements Contamination of Surface Soils in Shushtar and Dezful, Iran

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Abstract
Background: The potentially toxic elements (PTEs) are one of the most dangerous pollutants in the environment. In this study, the elements namely cadmium, lead, chromium, nickel, copper and zinc were investigated in the soil.
Methods: In this study, 144 composite samples were randomly prepared from surface soils in a depth of 10 cm in the cities of Shushtar and Dezful. Soil sampling was performed in 12 stations in Shushtar and Dezful. The PTEs were determined by ICP Varian 710-ES device.
Results: The pattern of accumulation of PTEs was as Cu > Pb > Cd > Zn > Ni > Cr in Dezful. In Shushtar, the accumulation of metals in the soil was as Cu > Pb > Cd > Ni > Zn > Cr. The concentration of Ni and Cr in the surface soils of Shushtar was higher than Dezful. The Ecological risk of Cd in the surface soils of Dezful and Shushtar was higher than other PTEs. The most important risk factor for carcinogenicity was related to Cr (3.15×10^{-7}) in children. Hazard quotient (HQ) value of studied PTEs for adults and children were obtained by ingestion, inhalation, and dermal contact absorption of less than 1.
Conclusion: According to the results, the PTEs of Cd, Pb and Cu caused high pollution in the soils of Shushtar and Dezful, which is due to agricultural, industrial and urban activities in these areas. In general, the metals Cr, Zn and Ni slightly contaminated the soil. Also, the ecological risk of PTEs showed that the highest effects on soil was related to Cd and Pb metals.
Keywords: Heavy metals contamination, Hazard quotient, Soil contamination, Toxic elements

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Introduction

Soil contamination is defined as presence, dispersal, or mixing foreign substances into the soil for a period of time by which the physical, chemical, and biological quality of the soil is altered, which is harmful to human, organisms, plants and buildings.[1-3] Soil contaminants can be solid or liquid that can be mixed with natural soil.[4,5] The main types of soil pollutants are oil contamination, industrial inputs, urban pollutants (sewage and waste) and agricultural pollutants.[6,7] The potentially toxic elements (PTEs) such as lead (Pb), cadmium (Cd), zinc (Zn), copper (Cu), herbicides and pesticides are the most important soil contaminants.[8] The most common metals derived from petroleum pollutants in the soil are Ni, V, Pb, Cr and Cd.[9,10] Vehicles accidents which produce contaminants are another example of soil contamination by human activities.[11] To spill toxic substances such as solvents, dyes and detergents can also spread soil.[12]

PTEs can have adverse effects on living organisms, aquatic animals and human.[13] It might also enter into human body through skin absorption and inhalation of dust, which indirectly damages human health through food contamination in urban areas.[14] The effects of PTEs on human health are different, though, but the main effect is appeared after entering into human body.[15] Due to increasing heavy metals, growth retardation, behavior change, neurological disorders, genetic change, toxicity of heavy metals, and accumulation in various organs of the human body, it is, therefore, important to study their concentrations.[16,17]

Fazekašová and Fazekaš examined the undesirable impurities of the deposit, mainly As, Pb and Zn in the Nizna Slana around a Fe mine, Slovakia. Based on the results, due to the environmental zoning of the Slovak, it can be concluded that the content of Mg, Cd, Cr, Cu, As, Fe, Mg and Mn is higher than the level of toxicity.[18] Rolka et al investigated the amount of PTEs in the peripheral soil along Sielska street in Olsztyn, Poland. The enrichment factor obtained for the Cd, Pb, Zn, Cu, Co and Ni might indicate their accumulation in the soil.[19] In addition, the study showed that proper land management including green belts, hedges and trees can reduce the PTEs transformation over long distances. Ennaji et al investigated the potential toxic metals named Cu, Zn, Cr, Cd, Pb and their aggregated impact on farm soils in the northeastern region of the Tadla Plain, Morocco. Principal component analysis (PCA) revealed no association among soil properties and PTEs content. The results showed that the metal elements are originated from several sources, human. Mean RI values indicated that there was a low ecological risk, and the study area is relatively contaminated with Zn and Cu which is partially affected by human activities.[20]

Based to JRC (Joint Research Centre Institute for Environment and Sustainability Soil and waste Unit) standard, the standard limits of cadmium, lead, nickel, chromium, copper and zinc in soil are 1.5, 100, 70, 100, 100 and 200 mg/kg, respectively.[21] The standard of cadmium, lead, nickel, chromium, copper and zinc is as 3.9, 300, 50, 0.4, 63 and 200 mg/kg, respectively based on Iran's national environmental standards.[22] Regarding the comparison of heavy metals with the permissible limits based on the standards, it should be stated that it does not show the severity of soil contamination, and it only shows whether the soil is contaminated with heavy metals or not. In fact, the thresholds of each metal are specific and the hazards of each metal are considered separately. Also, the permissible limits for heavy metals are variable in different regions, countries and applications.[23,24]

Therefore, because of wide using of land in Khuzestan province for urbanization, industrial activities, agriculture and mines the PTEs in the surface soils of different regions has been monitored. Also, to determine the PTEs status, human health risk assessment, ecological risk of the region and comparison of the PTEs concentration between the surface soils were the objectives of this study. Comparison of the heavy metals concentration in the surface soils in different areas of Shushtar and Dezful to the national and international standards as well as determination of the enrichment factor of the metals in the areas were other objectives of this study.

Materials and Methods

Study Area

This research was conducted in two regions of the northern part of Khuzestan province, Iran named Shushtar (48° 35′ and 32° 26′) and Dezful (48° 24′ and 32° 22′) (Figure 1). Shushtar and Dezful are important agricultural areas in Khuzestan province, which have been developed due to their agricultural prosperity, population growth, industrial development and urbanization. Khuzestan is one of the most important and valuable province of Iran.[25] There are abundant oil, gas, salt and sulfur mines in this area. According to the amount of raw material consumption and production quality, industries can be divided to two main categories named heavy industries and light industries. These mines include oil, gas, salt, sulfur, limestone, silica, gypsum and limestone.[10,26] Shushtar and Dezful are two important cities of Khuzestan province in terms of vegetables, summer crops, fruits and vegetables. In the industrial areas of the cities, there are various industries, including food and processing industries. Dezful is the second largest city in Khuzestan province after Ahvaz. Dezful includes four industrial towns which have workshops and chemicals factories, food industries, metals and rubber industries, such as

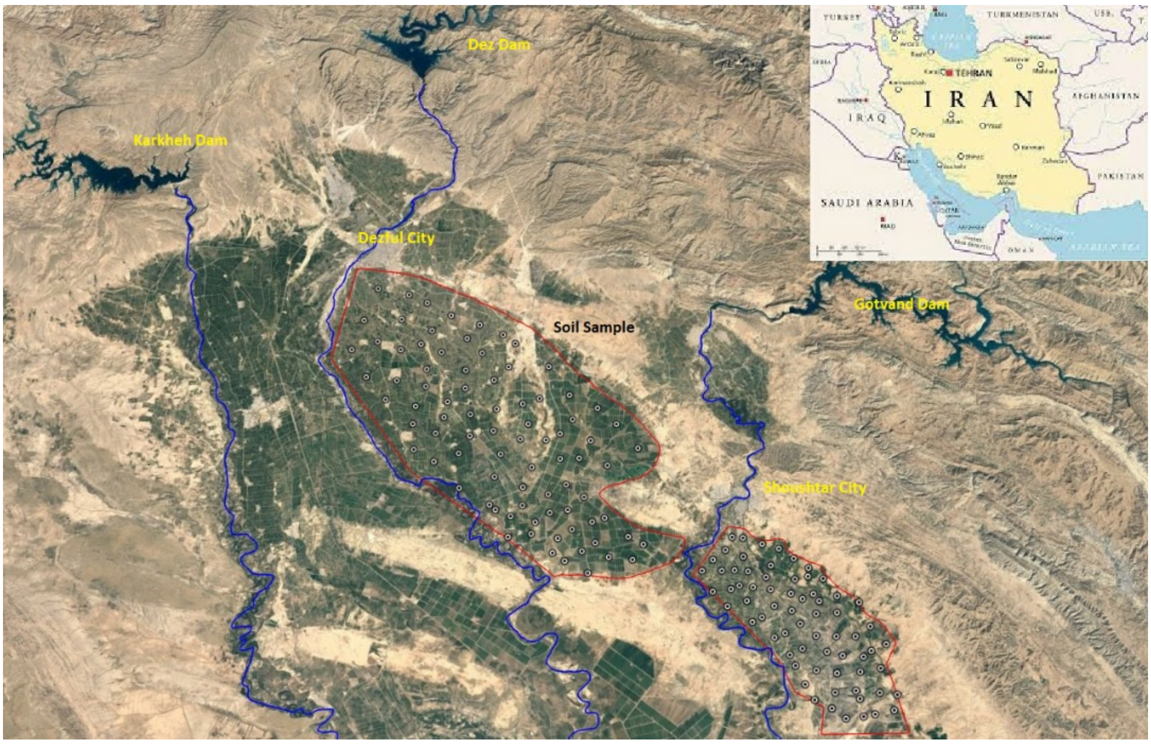

Figure 1. Location of Study Area and Soil Sample Locations in the Study Area.

Pars Polymer Elixir Company, Kooshan Food Industries and Dezful Aluminum. Shushtar has also two industrial towns with different industries such as Azin food industry factories, Khuzestan Golpooneh Company, Sabznam Food Industries Complex, Pars Petro Shushtar Company and Karun Agro-industry in Shushtar.

Soil Sampling and Measurement of PTEs

In this study, 144 composite samples were randomly prepared from surface soils to a depth of 10 cm from the cities of Shushtar and Dezful in the summer of 2020. Composite soil samples were considered as 1×1 m^2 plot. In other words, at each sampling point, an area of 1 square meter was considered and a soil sample was prepared from each vertex that were mixed together.

To evaluate sticking of heavy metals to major parts of soil, digestion approaches were used. It was defined as the process of Kimiapajoh Alborz Laboratory used in Shahr-e-Kord, in which 5 g of selected dusts from the street were mixed to soil samples using highly purified nitric-hydrofluoric per chloric acids (5 mL HNO–4 mL HF–$2HClO_4$), in order to disintegrate the selected units and measure the amount of potentially toxic metals, according to GB/T 17441-1997 and GB/T 17238-1997; USEPA, 1997).[27] Sulfur-hydrochloric-nitric-hydrofluoric acid approaches were applied to define Cr according to GB/T17137-1997.[28] The samples were pumped using a normal hydrochloric acid into a 50 cc balloon and injected into a pre-calibrated ICP Varian 710-ES device, and the amount of elements in each sample was then determined.[29]

The samples were washed using distilled water, and mud and soil were separated from them. Each sample was then individually labeled in Petri dishes and placed in an oven of 105°C for 48 hours. After drying and reaching a constant weight, the samples were crushed and sieved. 0.5 g of each sample was then weighed with a digital scale with an accuracy of 0.01 g. For chemical digestion, 0.5 g of each sample was poured into a 250 mL balloon, to which 25 mL of concentrated sulfuric acid, 20 mL of 7 M nitric acid and 1 mL of 2% sodium molybdate solution as well as a few boiling stones were added. It was placed in a location in which the boiling operation could be regular and uniform. After cooling the sample, 20 mL of a mixture of concentrated nitric acid and concentrated perchloric acid were slowly added to the sample from the top of the refrigerant in a ratio of 1:1. After then, we heat the mixture until the white vapors of the acid was completely disappeared. Ten mL of distilled water was slowly added to the cooled mixture from the top of the refrigerant while rotating the balloon. By heating (about 100 minutes), a completely clear solution was obtained, which was cooled and transferred to a 100 mL flask after cooling.

We evaluated quality assurance (QA) and quality control (QC) through assessment of blank and standard approach of NIST 2710. The approach showed a validity of $100 \pm 5\%$ ($n = 15$) while the precision of duplicate samples was between 4–6%. To ensure the data quality of the samples, standard approach of SRM 2710 was simultaneously used in 15% of the soil obtained from the surface soil.

Indications of Contamination

In order to evaluate the elements contamination in the top selected soils, contamination factor (CF) was applied (Equation 1). In Equation 1, Cn is the elements concentrated in the soil and Co represents the mean concentration of the element in the background. Based on the standards introduced by Hakanson, if $1 < CF$, $3 > CF \geq 1$, $6 > CF \geq 3$ and $6 \leq CF$, the pollution is at low, moderate, high and severe levels, respectively.[30]

$$\mathrm{Cf} = \frac{C_0}{C_n} \tag{1}$$

The sum of contamination factors for the studied elements indicates the degree of contamination (Cdeg) obtained from Equation 2. According to the classification of pollution levels, $8 < \mathrm{Cdeg}$ is considered as low pollution, $16 > \mathrm{Cdeg} \geq 8$ is considered as moderate pollution, $32 > \mathrm{Cdeg} \geq 16$ is considered as high pollution and $\mathrm{Cdeg} \geq 32$ is considered as severe pollution[30]:

$$\mathrm{Cdeg} = \sum \mathrm{Cf} \tag{2}$$

There were some limitations in Hakanson's degree of contamination index.[30] So, Abrahim presented a modified degree of contamination based on the CF of the contamination factor and the number of PTEs studied. Based on the classification of modified pollution levels, $1.5 < \mathrm{mCd}$, $2 > \mathrm{mCd} \geq 1.5$, $4 > \mathrm{mCd} \geq 2$, $8 > \mathrm{mCd} \geq 4$, $16 > \mathrm{mCd} \geq 8$ and $32 > \mathrm{mCd} \geq 16$ show no pollution, very low pollution, low pollution, moderate pollution, high pollution and extremely high pollution, respectively. In addition, $32 \leq \mathrm{Cdeg}$ indicates extremely high pollution in equation 3[31]:

$$\mathrm{mCd} = \sum \frac{\mathrm{Cf}}{\mathrm{n}} \tag{3}$$

The pollution load index (PLI) was estimated using equation 4. In the equation, CF represents the pollution factor, which is estimated through the association among the contamination factor of the metals. Number of metals has been presented by n in the formula. The range of PLI varies between zero (no contamination) to 10 (highly contamination). Specifically, the values less than 1 represent no contamination and those which are greater than 1 show contamination with PTEs[30]:

$$\mathrm{PLI} = \sqrt[6]{\mathrm{CF}_{n1} \times \mathrm{CF}_{n2} \times \ldots\ldots \times \mathrm{CF}_{n6}} \tag{4}$$

We estimated the enrichment factor of the metal through the ratio of normalized elements to the background values using equation 5. The element with purely geological source was considered as the reference element for the enrichment factor. In the present study, iron was considered as the reference metal. In order to estimate the concentration of PTEs in the soils, the samples were selected from pristine and untouched areas of Dezful and Shushtar. Based on suggested classification, $EF < 2$, $5 > EF \geq 2$, $20 > EF \geq 5$, $40 > EF \geq 20$ and $40 \leq EF$ show low, moderate, high, very high and extremely high level of pollution[30]:

EF = (Metal / Reference metal) Sample ÷ (Metal / Reference metal) Background (5)

Ecological risk assessment and biological potential risk index (RI) of the selected soils were calculated using equations 6-7. In this regard, CF is pollution factor; Er indicates the ecological risk of each element; RI represents the ecological risk of the total elements. Hakanson defined the TR which is an indicator of the PTEs toxicity. Ecological risk for each element is classified into five levels including $40 < Er$, $80 > Er \geq 40$, $160 > Er \geq 80$, $320 > Er \geq 160$ and $Er \geq 320$ which represent low risk, medium risk, significant risk, high risk and very high risk, respectively. To analyze the ecological RI, four categories of low ecological risk ($RI < 150$), medium ecological risk ($300 > RI \geq 150$), significant ecological risk ($600 > RI \geq 300$) and very high ecological risk ($RI \geq 600$) are classified in the following equations.[30]

$$Er = TR \times Cf \tag{6}$$

$$RI = \sum E_r \tag{7}$$

Geo-accumulation indicator was first developed by Muller which estimate the soil contamination degree. It is applied in environmental sciences to indicate contaminated surfaces. The index is estimated using equation 8, in which Igeo represents the land accumulation index, Cn is the PTEs concentration of soil, Bn represents the concentrated average shale in the ground. To address the effects of soil materials and natural fluctuations in the content of the given substance in the environment and for the very little change caused by human activities, a coefficient of 1.5 was used. Muller classification includes seven classes of contamination; Igeo: < 0, Igeo :1-0, Igeo: 1-2, Igeo: 2-3, Igeo: 3-4, Igeo: 4-5 and Igeo < 5 which represent healthy, healthy and slightly infected, slightly infected, slightly infected to highly infected, highly contaminated, highly contaminated to severely infected and highly classified soil, respectively.[32]

$$I_{geo} = \mathrm{Log2}\left(Cn / 1.5 \times Bn\right) \tag{8}$$

Health Risk Assessment

The PTEs were treated according to the health risk assessment approach introduced through the US Environmental Protection Agency (USEPA). This assessment was divided to two parts; carcinogenic and non-carcinogenic risks of exposure to metals through three pathways namely swallowing, respiration and skin absorption. Mean daily dose was calculated using Equations 9-11 in each route.[33]

$$ADD_{ing} = \frac{C \times IngR \times CF \times EF \times ED}{BW \times AT} \tag{9}$$

$$ADD_{inh} = \frac{C \times InhR \times EF \times ED}{PEF \times BW \times AT} \tag{10}$$

$$ADD_{dermal} = \frac{C \times SA \times CF \times AF \times ABF \times EF \times ED}{BW \times AT} \tag{11}$$

In these equations, ADD_{ing}, ADD_{inh} and ADD_{dermal} represent the mean daily absorption of metals (mg/kg/day) by ingestion, inhalation, and dermal absorption, respectively. Also, C represents concentrated metals of the soil (mg/kg). Swallowing and soil respiration rates (mg/day and m^3/day) are shown by IngR and InhR, respectively. In addition, EF is the frequency of exposure to metal (day/year), ED represents duration of exposure to metals (year); BW is weight of the person exposed to metals (kg); AT represents time of exposure to average of all metals (day); CF is the conversion factor(m^3/kg); SA represents the size of skin exposed to metals (cm^2); AF is the soil adhesion factor (mg/cm^2/day) and ABF is the skin adsorption factor (unit). Non-carcinogenic risk (HI) of the entire ingestion, inhalation, and dermal absorption pathways for young and elderlies was identified using the total daily absorption of PTEs (ADD) in each pathway to the reference value of that metal toxicity by equation 12.[33]

$$HQ_i = \sum \frac{ADD_i}{R_f D_i} \tag{12}$$

In this regard, HQ is the risk of pathway-specific non-carcinogenicity of metals; ADDi is the pathway-specific daily absorbed metal (mg/kg-day). The HQ of less than 1shows that it is compatible with human health. The HQ of higher than 1shows that it has adverse and worrying impact on health. The value of total non-carcinogenic hazard (HI) of all metals in young and elderlies was estimated based on equation 13[33]:

$$HI = \sum HQ_i \tag{13}$$

Assessment of the carcinogenic risk of each of the three pathways for these metals was performed using equation 14[33]:

$$RI = \sum ADD_i \times SF_i \tag{14}$$

In the high ratio of carcinogenic risk (RI), ADDi of daily metals uptake values in each metal exposure pathways (mg/kg/day) and SF_i carcinogenicity factor per unit of metal exposure (mg/kg/day).

Measurement of Physical and Chemical Parameters

The acidity (pH) of the soil was measured using the samples prepared according to ISO10390 standard method and pH meter, Model 691. To determine the electrical conductivity (EC), the sample was prepared according to the ISO11265 standard method, and the electrical conductivity of the sample was measured by an EC meter.[34] Determination of soil texture is done using hydrometric method. Also, the soil organic carbon was measured by the walkie-talkie method.[35]

Transfer Factor (TF)

The total concentration ratio of HMs in vegetables to the total concentration of HMs in the soil is defined as transferring factor. It helps evaluate the plants' ability of HMs transformation from soil to their edible parts. It was calculated by equation 15[36]:

$$\text{Transfer factor (TF)} = C_{vegetable}/C_{soil} \qquad (15)$$

In which the $C_{vegetable}$ and C_{Soil} represent concentration of HMs in the edible vegetable component (mg/kg) and its soil, respectively. The transformation factor of higher than 1 shows that there is a significant cumulative concentration of HMs in the vegetable.[37]

Statistical Analysis

The data were analyzed using SPSS 24 statistical software. Excel 2007 software was also used to calculate the contamination indices. Pearson's correlation and cluster analysis were used to investigate the relationship between PTEs data and detect metals, respectively.

Results and Discussion

Statistical parameters of PTEs, pH, EC, soil organic carbon (SOC), including kurtosis and skewness values in surface soils of Dezful and Shushtar are presented in Table 1. The pattern of accumulation of PTEs in the soil of Dezful was as Cu > Pb > Cd > Zn > Ni > Cr. In Shushtar, the accumulation of metals in the soil was Cu > Pb > Cd > Ni > Zn > Cr. The content of Cd, Pb, Zn and Cu in the surface soils of Dezful was higher than Shushtar. The concentration of Ni and Cr in the surface soils of Shushtar was higher than Dezful. A research reported that accumulation of PTEs in the surface soils of Ahvaz was Cu > Pb > Cr > Zn > As > Cd and the high concentrations in addition to a high coefficient of variation indicated the human role for toxic metals.[26] Guan et al investigated the amount of PTEs in an industrial and mining area called Tianjin in China. They found that the highest levels of contamination in the study area were related to Zn, and the contamination of Cu, Cd and Pb was higher than average earth's crust.[38] Adesuyi et al conducted a study in industrial areas of Nigeria in soil samples from twenty-two areas. They measured the Zn, Cu, Pb, Fe and Cd on the samples. The highest contamination was related to zinc with a value of 141.06 mg/kg and Cu with a concentration of 131.7 mg/kg soil.[39] Mining activities cause soil erosion as a result of environmental contact with a wide range of PTEs.[40,41] The PTEs in the soil can vary depending on the soil layer and class. Also, the type and amount of chemical fertilizers that are added to the soil affect the accumulation of PTEs in the soil.[42] While the Ni, Zn and Cr contamination factor in surface soils of Dezful and Shushtar showed low pollution, but Pb and Cu had moderate pollution. Regarding Cd, the pollution factor showed low pollution in the soils of Shushtar and Dezful city. The results of three important indicators including degree of pollution, degree of modified pollution and load index of pollution PTEs in the soils of Dezful and Shushtar showed that these pollutants had little effect on the soil, and caused very little pollution. The PTEs

Table 1. Concentration of PTEs (mg/kg) in Surface Soils in Dezful and Shushtar

Location	PTEs	Min	Max	Mean	Standard Deviation	Standard Error	Variance	Skewness	Kurtosis	CV
	Cd	0.46	2.56	1.43	0.77	0.18	0.599	0.151	-1.643	0.53
	Pb	2.34	97.50	32.90	31.04	7.31	963.68	1.002	-0.068	0.94
	Ni	0.01	0.89	0.26	0.31	0.07	0.097	1.133	-0.256	1.19
	Cu	30	131.12	74.34	36.0	8.62	1339.85	0.418	-1.491	0.48
Dezful	Zn	0.11	0.59	0.27	0.18	0.04	0.03	0.822	-1.253	0.66
	Cr	0.03	0.10	0.05	0.02	0.005	0.001	0.594	-1.516	0.4
	pH	7.20	7.56	7.39	0.11	0.02	0.01	-0.258	-1.117	0.01
	Ec (ds/m)	0.38	0.81	0.58	0.13	0.03	0.01	0.231	-0.684	0.002
	SOC (%)	0.25	0.71	0.46	0.15	0.03	0.02	0.349	-1.311	0.32
	Cd	0.22	1.76	0.71	0.05	0.11	0.244	1.439	0.927	0.07
	Pb	2.11	42.18	18.90	13.50	3.18	182.35	0.404	-1.307	0.71
	Ni	0.01	1.85	0.51	0.66	0.15	0.44	1.110	-0.239	1.29
	Cu	12.50	115.76	67.41	34.57	8.14	1195.41	-0.152	-1.225	0.51
Shushtar	Zn	0.1	0.39	0.19	0.01	0.02	0.01	0.923	-0.588	0.05
	Cr	0.01	0.45	0.08	0.01	0.02	0.01	1.132	1.332	0.12
	pH	7	7.71	7.42	0.22	0.05	0.052	-0.756	-0.740	0.02
	Ec (ds/m)	1.06	4.71	2.42	1.52	0.36	2.338	0.655	-1.531	0.62
	SOC (%)	0.1	0.29	0.19	0.06	0.01	0.005	0.306	-1.393	0.31

naturally exist in the earth's crust and, despite their low content and low solubility, they are generally separated from the earth's crust by weathering and erosion and enter into ecosystems. Human-made pollution enters into soil, which aquatic ecosystems and increases the amount of PTEs in these environments, leading to many problems for animals and plants.[43,44] The average concentrations of arsenic, cadmium, chromium, copper, lead and zinc in the soils of Eghlid city in southern Iran were 1.85, 2.80, 19.04, 19.35, 7.17 and 38.77 mg/kg, respectively.[11] Another study conducted in the surface soils of industrial town No. 2 in Ahvaz reported the concentration range of heavy metals cadmium as 6.70-12.64 mg/kg. It also reported that vanadium, iron and copper ranged between 23.14-50.12 mg/kg, 18980-22456 mg/kg and 31.85-42.46 mg/kg, respectively.[45] Many studies have showed that the concentration of heavy metals in topsoil can vary.[10,11,16,26,45] This difference in concentration of heavy metals may be due to human activities such as industry, agriculture and urbanization. However, the geological processes of the region can also affect the concentration of heavy metals in the soil.[8,20]

The contamination factor (Cf) values of Cu (2.25), Cd (7.15) and Pb (1.94) in the surface soils of Dezful was higher than Shushtar. However, Cf of Cr (0.0008) and Ni (0.03) metals was higher in Shushtar soil. The EF values of Zn (0.44), Cr (0.03), Cd (315.99) and Cu (99.75) in Dezful surface soils was higher than Shushtar, but the Pb (92.08) and Ni (0.24) EF was higher in Shushtar. The Igeo values of Cu (0.41), Cd (2.02), Zn (-7.51) and Cr (-11.41) in Dezful surface soils was higher than Shushtar. The values of Pb (0.18) and Ni (-8) metals was higher in Shushtar soil. The Cdeg and mCd of PTEs pollution of surface soils of Dezful city were higher than Shushtar city. The Cfs of PTEs in soil of Dezful were as Cd > Cu > Pb > Ni = Zn > Cr. The index of soils for Shushtar was aa Cd > Cu > Pb > Ni > Zn > Cr. The pattern of Igeo and EF in the soil of Dezful city was as Cd > Cu > Pb > Zn > Ni > Cr. The values of these two indices for PTEs in the soils of Shushtar city were as Cd > Pb > Cu > Zn > Ni > Cr (Table 2). The results of three important indicators including degree of pollution, modified degree of pollution and load index of heavy metal pollution in surface soils of Dezful and Shushtar were less than 1. These results showed that these pollutants had little effect on soil, and caused very little pollution. Because the value of the contamination load index of all selected metals was more than 1, showing higher number of contamination sites than the non-contaminated ones).[46] In other words, compared to the background concentration, the contamination load index shows that the period of heavy metals concentration of soil increased which indicates a brief toxicity of the selected metals.[47] Heavy metals naturally exist in the earth's crust and, despite their low content and low solubility, they are generally separated from the earth's crust by weathering and erosion and enter into the ecosystem. Human-made contaminants enter into soils and aquatic ecosystems which increase the amount of heavy elements in these environments, causing many problems for animals and plants.[26,43]

Table 2. Environmental Indicators of PTEs in Surface Soils in Dezful and Shushtar

Location	PTEs	CF	EF	Igeo	Cdeg	mCd	PLI
Dezful	Cd	7.15	315.99	2.02	11.36	1.89	0.22
	Pb	1.94	85.97	-0.46			
	Ni	0.01	0.12	-8.01			
	Cu	2.25	99.75	0.41			
	Zn	0.01	0.44	-7.51			
	Cr	0.0005	0.03	-11.41			
Shushtar	Cd	3.75	161.34	0.98	6.94	1.15	0.23
	Pb	1.11	92.08	0.18			
	Ni	0.03	0.24	-8			
	Cu	2.04	49.98	-0.91			
	Zn	0.01	0.32	-7.88			
	Cr	0.0008	0.02	-12.32			

CF: Contamination factor; EF: Enrichment Factor; Igeo: Geoaccumulation index; Cdeg: degree of contamination; mCd: modified degree of contamination; PLI: pollution load index.

The EF value of Ni, Zn and Cr showed that the amounts of these metals in the soils of the two regions were the result of natural erosion and geological and anthropogenic activities did not play an effective role in the mineralization of these metals. However, very high concentration of Cd in the study areas might be due to industrial, agricultural and urban resources. High concentration of Pb and Cu was also observed, which are influenced by anthropogenic activities. Among the most important artificial sources of soil pollution and consequently Cd in agricultural products, the discharge of industrial wastewater sludge, the use of superphosphate fertilizers and the burial of non-ferrous waste in the ground and the location of agricultural lands are limited to Pb and Zn mines or refineries.[48,49] Shushtar city has gypsum mines, lime and building stone as well as sand. Shushtar includes two industrial towns and different industries such as Azin food industry factories, Khuzestan Golpooneh Company, Sabznam Food Industries Complex, Pars Petro Shushtar Company and Karun Agro-industry in Shushtar. Dezful city includes four factories and industrial towns such as workshops and factories of chemicals, food industries and metal and rubber industries such as Pars Polymer Elixir Company, Kooshan Food Industries, and Dezful Aluminum. Bhuyan and Islam examined heavy metal contamination in Bangladesh's agricultural soils using several indicators, including enrichment factor (CF), Accumulation Index (Igeo) and PLI. The results showed that the soil was significantly enriched with titanium, manganese, zinc, lead, arsenic, iron, strontium and antimony as a result of mineral activity.[50] Salman et al evaluated heavy metal contamination in Arabi farms in Elbor, Egypt. Soil contamination with these metals was calculated using pollution factors, degree of pollution, PLI, environmental risk factor (Er), potential environmental risk index and land accumulation index. Integrated pollution calculation indices namely PLI, pollution degree and potential environmental risk index showed that the observed soil were infected with the heavy metals. extreme concentration of arsenic and cadmium showed that they play an important role on the high concentration of soil pollution.[6] The Er values of PTEs in the surface soils of Dezful were as: Cd > Pb > Cu > Ni > Zn > Cr. However, the Erin of surface soils of Shushtar was different (Cd > Cu > Pb > Ni > Zn > Cr). The ecological risk of Cd in the surface soils of Dezful and Shushtar was higher than other PTEs. The total risk index of PTEs in the surface soils of Dezful was higher than Shushtar (455.55 vs 123.10) (Table 3). A study revealed that the risk index of heavy metals in surface soils of Ahvaz city was 157.42, which is at a moderate level and consistent with the results of this study.[26] The ecological risk of heavy metals indicates the status of heavy metal pollution in the soil of the region.[11,30] According to the values derived from this index, we can say that the surface soils of Dezful have higher pollution than heavy metals.

Human Health

Results of human health risk assessment indicated that the daily absorption rate of PTEs by ingestion in children was higher than inhalation and dermal contact absorption. The highest daily absorption of copper was in children and adults (Table 4). Hazard quotient (HQ) values of PTEs for adults and children were obtained by ingestion, inhalation and dermal contact absorption less than 1 (Table 5). The carcinogenicity index of Cd, Pb, Ni and Cr for adults and children was less than 10^{-4}. The highest risk factor for carcinogenicity was related to Cr metal (3.15×10^{-7}) in children. The non-carcinogenic risk index of Pb metal in the soils of Shushtar (0.069) and Dezful (0.121) was higher than other PTEs (Table 6). The PTEs health risk assessment showed that the risk index and carcinogenic risk index of metals were less than 1 and $10^{-4,}$ respectively. In consistent with these results, the PTEs health risk assessment in Bangladesh's Tangail Industrial Zone indicated that a total hazard index for each concentrated metal was below 1, and cancer risk values were lower than 10^{-4}, leading to a lack of cancer or low risk of cancer in adults and young people.[51] Yang et al examined the status and pattern of potential health hazards from the accumulation of metals in agricultural soils in China. In this study, PTEs, As, Cr, Ni, Cd, Pb and Zn were selected as pollution and hazard assessment. The Cd concentrations with the highest mean accumulation index were higher than their reference standard. In addition, the mean risk index (HI) of exposure to six PTEs had non-cancerous risks for children over 1 year. Arsenic was extremely high for cancer and non-cancer risks, while both Cr and Cd were high-risk metals with high cancer risk.[52] Also, the assessment of soil heavy metals for human health in China's Yangtze Basin region has shown that children, regardless of their carcinogenic or non-carcinogenic risk, are prone to potential metal-related health risks. While there were no significant carcinogenic and non-carcinogenic risks in adults, but a significant non-

Table 3. Ecological Risk assessment of PTEs in surface soils in Dezful and Shushtar

Location	PTEs	Ecological Risk	Risk Index
Dezful	Cd	214.5	455.55
	Pb	147.5	
	Ni	0.2	
	Cu	93.3	
	Zn	0.04	
	Cr	0.01	
Shushtar	Cd	107.17	123.10
	Pb	5.56	
	Ni	0.13	
	Cu	10.21	
	Zn	0.01	
	Cr	0.02	

Table 4. Average Daily Dose of PTEs in Surface Soils of Shushtar and Dezful

Location	Metals	ADD_{ing}		ADD_{inh}		ADD_{dermal}	
		Adult	Children	Adult	Children	Adult	Children
Dezful	Cd	1.82×10^{-4}	2.45×10^{-5}	5.12×10^{-9}	2.30×10^{-9}	2.92×10^{-7}	7.46×10^{-7}
	Pb	0.0004	5.64×10^{-4}	1.17×10^{-7}	5.31×10^{-8}	6.73×10^{-6}	1.71×10^{-5}
	Ni	3.32×10^{-5}	4.46×10^{-6}	9.32×10^{-10}	4.19×10^{-10}	5.31×10^{-8}	1.35×10^{-8}
	Cu	0.0009	0.0001	2.66×10^{-7}	1.20×10^{-7}	1.52×10^{-5}	3.88×10^{-5}
	Zn	3.45×10^{-5}	4.63×10^{-6}	9.68×10^{-10}	4.35×10^{-10}	5.52×10^{-8}	1.41×10^{-7}
	Cr	6.39×10^{-6}	8.57×10^{-7}	1.79×10^{-10}	8.07×10^{-11}	1.02×10^{-8}	2.61×10^{-8}
Shushtar	Cd	9.07×10^{-5}	1.21×10^{-5}	2.54×10^{-9}	1.14×10^{-9}	1.45×10^{-7}	3.70×10^{-7}
	Pb	0.0002	3.24×10^{-4}	6.77×10^{-8}	3.05×10^{-8}	3.86×10^{-6}	9.87×10^{-6}
	Ni	6.52×10^{-5}	8.74×10^{-6}	1.82×10^{-9}	8.23×10^{-10}	1.04×10^{-7}	2.66×10^{-7}
	Cu	0.0008	0.0001	2.41×10^{-7}	1.08×10^{-7}	1.37×10^{-5}	3.52×10^{-5}
	Zn	2.42×10^{-5}	3.25×10^{-6}	6.81×10^{-10}	3.06×10^{-10}	3.88×10^{-8}	9.92×10^{-8}
	Cr	1.02×10^{-5}	1.37×10^{-6}	2.86×10^{-10}	1.29×10^{-10}	1.63×10^{-8}	4.17×10^{-8}

ADD_{ing}: ingestion, ADD_{inh}: inhalation and ADD_{dermal}: dermal contact absorption.

Table 5. Hazard Quotient of PTEs in Surface Soils of Shushtar and Dezful

Location	Metals	ADD_{ing}		ADD_{inh}		ADD_{dermal}	
		Adult	Children	Adult	Children	Adult	Children
Dezful	Cd	0.060	0.008	1.65×10^{-5}	7.44×10^{-6}	0.0002	0.0006
	Pb	0.120	0.016	3.35×10^{-5}	1.50×10^{-5}	0.001	0.003
	Ni	0.003	0.0004	9.32×10^{-7}	4.19×10^{-7}	0.0005	0.001
	Cu	0.023	0.003	6.63×10^{-6}	2.98×10^{-6}	0.0001	0.0003
	Zn	1.15×10^{-4}	1.54×10^{-5}	3.22×10^{-9}	1.45×10^{-9}	9.20×10^{-7}	2.35×10^{-6}
	Cr	0.0002	2.85×10^{-4}	6.27×10^{-6}	2.82×10^{-6}	1.70×10^{-4}	4.35×10^{-4}
Shushtar	Cd	0.030	0.004	8.21×10^{-6}	3.69×10^{-6}	0.0001	0.0003
	Pb	0.069	0.009	1.92×10^{-5}	8.66×10^{-6}	0.0007	0.001
	Ni	0.006	0.0008	1.829×10^{-6}	8.23×10^{-7}	0.001	0.002
	Cu	0.021	0.002	6.01×10^{-6}	2.70×10^{-6}	0.0001	0.0002
	Zn	8.09×10^{-5}	1.08×10^{-5}	2.27×10^{-9}	1.02×10^{-9}	6.47×10^{-7}	1.65×10^{-6}
	Cr	0.0003	4.57×10^{-4}	1.003×10^{-5}	4.51×10^{-6}	2.72×10^{-4}	6.96×10^{-4}

Table 6. Evaluation of Carcinogenic Risk Index (RI) and Non-carcinogenic Risk Index (HI) of PTEs in Surface Soils of Shushtar and Dezful

Location	Metals	RI		HI	
		Adult	Children	Adult	Children
Dezful	Cd	2.70×10^{-7}	2.01×10^{-7}	0.008	0.061
	Pb	1.74×10^{-8}	1.29×10^{-7}	0.019	0.121
	Ni	2.06×10^{-8}	1.53×10^{-7}	0.001	0.003
	Cu	-	-	0.003	0.023
	Zn	-	-	1.77×10^{-5}	1.15×10^{-4}
	Cr	2.64×10^{-8}	1.97×10^{-7}	7.24×10^{-4}	0.0002
Shushtar	Cd	1.34×10^{-8}	1.001×10^{-7}	0.004	0.030
	Pb	1.001×10^{-8}	7.46×10^{-8}	0.011	0.069
	Ni	4.05×10^{-8}	3.02×10^{-7}	0.0035	0.0075
	Cu	-	-	0.0031	0.0216
	Zn	-	-	1.25×10^{-5}	8.16×10^{-5}
	Cr	4.23×10^{-8}	3.15×10^{-7}	0.0001	0.0003

carcinogenic effect was observed in children.[53] Based on the standard developed by US Environmental Protection Agency, if the daily metal absorption (EDIi) is higher than the reference point (RFDi), the risk of pathway-specific non-carcinogenic metals will be higher than the permittable limit (HQ > 1) which has adverse and worrying effects on human health.[54] The reference daily intake of cadmium, lead, zinc and copper is 0.001, 0.3, 0.02 and 0.04 mg/kg/day, respectively.[55] It should be noted that, in this study, the total daily intake of chromium, copper, zinc, arsenic, lead and cadmium in humans was less than the tolerable daily intake provided by the US Environmental Protection Agency.

The results of Pearson correlation showed that there was no correlation between most variables (Table 7), and there was only a positive and significant correlation between Cu and Pb ($R = 0.704$; $P < 0.01$), Cu and Ni ($R = 0.674$; $P < 0.01$) and Zn and Cd ($R = 0.827$; $P < 0.01$). In this study, Pearson correlation, PCA and cluster analysis of PTEs, PH, EC and SOC showed that there was no significant and positive correlation between variables. High correlation between metals and parameters indicates that the source of PTEs pollution is human activities. However, the lack of correlation between data and parameters indicates that their origin is natural and geological processes in the study area. Based on cluster analysis of PTEs and physical and chemical parameters in surface soils of Dezful and Shushtar cities, they were divided into two main groups. The first group includes Ni, Cd, Zn, Pb and Cr along with pH, EC and SOC. The second group consisted of Cu. The different cluster groups represent differences in the geochemical behavior and different origins of metals. Therefore, the metals in a group had the same natural origin (Figure 2). PCA (PCA) showed that there was a positive correlation in the first component, which includes Pb, Cu, Cr and Ni. In the second component, a positive and significant relationship was found between Cd and Zn soil. In the second component, pH had a negative correlation with other soil parameters. Soil organic carbon in the third component had a significant relationship with PTEs, and EC had a negative correlation in all three components (Table 8). According to cluster analysis in this study, it can be said that the origin of lead and copper metals was different from other heavy metals. It is also possible that the trend of heavy metal pollution in the study areas is related to copper and lead metals, and it seems that these metals will play a decisive role in soil heavy metal pollution in the future. According to the cluster and correlation analysis of heavy metals, chromium, cadmium,

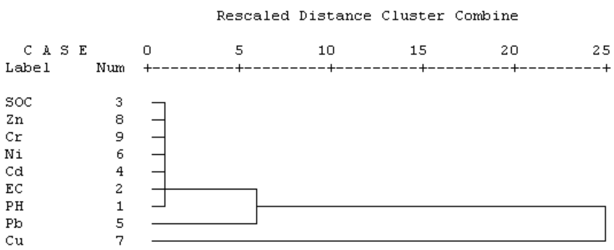

Figure 2. Cluster Analysis of PTEs and Physical and Chemical Parameters in Surface Soils of Shushtar and Dezful

Table 7. Pearson Correlation of PTEs, pH, SOC and EC in Surface Soils of Shushtar and Dezful

	PH	EC	SOC	Cd	Pb	Ni	Cu	Zn	Cr
pH	1								
EC	0.110	1							
SOC	0.071	-0.343*	1						
Cd	-0.298	-0.057	0.443**	1					
Pb	0.193	-0.336*	0.052	-0.331*	1				
Ni	0.198	-0.157	-0.402*	-0.408*	0.415*	1			
Cu	0.326	-0.269	-0.174	-0.036	0.704**	0.674**	1		
Zn	-0.536**	-0.025	0.242	0.827**	-0.452**	-0.389*	-0.228	1	
Cr	0.040	-0.248	-0.081	-0.132	0.387*	0.320	0.356*	-0.131	1

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

nickel and zinc metals probably have different sources and it seems that the low pollution of these metals is due to low urban activities. The source of PTEs can be man-made activities or natural processes.[7,53] The PTEs are the result of erosion processes, chemical and physical reactions of geology and weathering of rocks,[10] and human resources include agricultural, industrial and urban life activities.[56] The correlation between metallic elements and main oxides in nature is managed through some main factors like hotspots and soil geochemical characteristics of the study area, which distinguishes the terrestrial pattern from human.[57] In uncontaminated soils, main oxides like calcium, magnesium, iron, and potassium are the major controlling factors in the trace element distribution pattern.[58]

The average amounts of PET_S in the studied summer crops are listed in Table 9. The highest values of TF in Dezful and Shushtar were related to zinc metal (111.51 and 38.68). The lowest values of this factor were 0.03 and 0.02 for Pb metal (Table 9). The average of Pb, Cd and Cu in top soils (32.90, 1.43 and 74.34 mg/kg in Dezful and 18.90, 0.71 and 67.41 mg/kg in Shushtar) were higher than the average values of the earth's crust (Table 9). The concentrations of Cr, Zn and Ni were lower than the mean of the earth's crust, indicating that the Pb, Cd and Cu from anthropogenic activities have entered into the soils of the areas. Industries, such as plating and painting metal, soldering, rubber and plastics industries as well as urban and agricultural activities significantly affect the total Cd concentration.[59,60] Also, regardless of atmospheric deposition, the most important route for Cd to enter the urban dust is phosphorus fertilizers on agricultural lands within and around cities. The Cd also pollutes the environment by car tire wear and tear.[61] The impact of urban activities and industrial uses is one of the reasons for the high level of Pb in the soil of this region. Fossil fuels, coal fuels, vehicle traffic, and red tape coatings are also factors that increase Pb in the soil.[62] The Cu is used in industries for the production of copper pipes, cables, wires, and copper cookware. The most important sources of Cu for environmental pollution are mining, agriculture and waste and sewage treatment sludge.[63,64] Small amount

Table 8. Component Score Coefficient Matrix of PTEs and soil parameters of Dezful and Shushtar

	1	2	3
pH	0.046	-0.782	0.181
EC	-0.557	-0.114	-0.501
SOC	-0.098	0.044	0.937
Cd	-0.113	0.741	0.456
Pb	0.756	-0.371	0.148
Ni	0.697	-0.233	-0.438
Cu	0.849	-0.157	-0.039
Zn	-0.183	0.900	0.208
Cr	0.652	0.038	-0.066

Extraction Method: Principal Component Analysis.
Rotation Method: Varimax with Kaiser Normalization.

Table 9. Comparison of PTEs Concentration (mg/kg) in Surface Soils, Earth's Crust, Crops and TF in Dezful and Shushtar

PTEs	Dezful			Shushtar			earth's Crust[76]
	Soil	Crops	TF	Soil	Crops	TF	
Cd	1.43	0.41	0.28	0.71	0.25	0.35	0.09
Pb	32.90	1.18	0.03	18.90	0.44	0.02	15
Ni	0.26	0.04	0.15	0.51	0.03	0.05	47
Cu	74.34	23.23	0.31	67.41	4.16	0.06	29
Zn	0.27	30.11	111.51	0.19	7.35	38.68	31
Cr	0.05	1.31	26.20	0.08	0.22	2.75	35

of copper is essential for humans, though, if it increases, it is dangerous for human health. The maximum allowable daily intake of copper for adults is 0.9 mg per day.[65] The amount of Zn in the surface soils of Shushtar and Dezful was much lower than the average earth crust (75 mg/kg). The Zn content in natural soil is related to the chemical composition of the parent rock and the rate of weathering processes.[66] In agricultural soils, Zn is not even distributed and its content range is between 10 and 300 mg/kg.[67,68] Researchers believe that the normal Zn content in contaminated soils is very different ranging from 10 to 100 mg/kg.[69] The Zn deficiency in soils has been reported from several parts of the world. In a comprehensive study by the FAO, more than 30% of the studied soils were suffered from zinc deficient. The report revealed that Zn deficiency is more severe in some countries, including Belgium, Malta, Iraq, Turkey, Pakistan, and India than Syria, Lebanon, Mexico, and Thailand.[24]

Although the TF of Cd, Pb, Ni and Cu was less than 1 in this study, but this specificity for Zn and Cr in agricultural products of Dezful and Shushtar was higher than 1, showing high bioaccumulation potential for Zn and Cr. Environmental factors are effective for the absorption and accumulation of heavy metals in the straw plant.[70] The adsorption of a metal can also be affected by the presence of other inorganic elements and toxic metals through competition at the adsorption site.[71] The value less than 1 for the transfer factor indicates a greater tendency of the plant to accumulate the element in terrestrial organs than plant organs. Element solubility in tissue fluids and metabolism of plant species are among the main factors determining the values of this parameter.[72,73] Cd is an unnecessary and toxic element for plants which is rare in nature and the environment. Ni is a group of trace elements that have toxic effects on plants and has a slow and passive property in sediments.[74,75]

Conclusions

According to the results, the PTEs of Cd, Pb and Cu caused high pollution in the soils of Shushtar and Dezful, which might be due to agricultural, industrial and urban activities in these areas. In general, the Cr, Zn and Ni metals were slightly contaminated in the soil. The ecological risk of PTEs also showed that the highest effect on soil was related to Cd and Pb metals. PTEs health risk assessment showed that the risk index and carcinogenic risk index of metals were less than 1 and 10^{-4}, respectively, according to which, PTEs in the soil do not cause poison in human. However, due to high enrichment of cadmium, lead and copper in the soil of these areas, it is recommended that we should monitor PTEs in the soil, plants and biological indicators of Dezful and Shushtar regions continuously.

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Competing Interest

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