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ORIGINAL PAPER

Analysis and potential ecological risk assessment of heavy metals in the surface soils collected from various land uses around Shazand Oil Refinery Complex, Arak, Iran

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Abstract

Due to the presence of organic compounds, heavy metals, various hydrocarbons, organic solvents, aromatic compounds, linear formaldehyde, fats, and grease, petroleum products are among the most life-threatening factors in the ecosystem. Therefore, this study was conducted to examine the soil surrounding Shazand Oil Refinery Complex in Markazi Province, Iran, and assess the concentration of various heavy metals, including Fe, Cd, Mn, Cr, Zn, Cu, Ni, and Pb (mg/kg) in surface soils of different land uses. In so doing, the soil contamination rates and potential ecological risks of the soils were assessed using I_{geo}, IPI, EF, PLI, NIPI, and PERI. Based on the results of the I_{geo} index, moderate levels of Cd and Pb concentration (0.633 and 0.921, respectively) were observed. Based on the NIPI values, Cd (1.65) and Pb (2.01) could be classified as causing moderate pollution levels. Moreover, based on its E_i value, Cd (69.8) could be considered as posing a moderate ecological risk. Besides, the EF values of Cd (2.69), Pb (2.02), and Zn (1.41) indicated that they have minor enrichment. Besides, the IPI of the studied soil samples suggested that the soils could be categorized as low polluted, while the pollution load index (PLI) indicated that the whole research area could be considered as non-polluted. Based on the Pearson correlation coefficient, PCA, and HCA, it was decided that the heavy metals in the study area mainly originate from three different sources. Moreover, no significant difference was found between different land uses regarding the contamination of surface soil samples. However, in the long term, due to the impact of anthropogenic activities, discharge of Cd, Pb, and Zn to the environment might result in their accumulation in soil. In conclusion, it is suggested that soil analyses be included in future studies for determining the impact of the number of bioavailable metals.

Keywords Heavy metals · Pollution indices · Potential ecological risk · Shazand Oil Refinery Complex · Soil contamination

Introduction

Soil is the most abundant biomaterial on earth and one of the foundations of human life, providing some essential ecosystem services. Obviously, human activities such as the misuse of land or its pollution disturb this ecosystem service, and this can be harmful to runoffs, underground water resources, sediments, oceans, and organisms (Adhikari and Hartemink 2016; Schaeffer et al. 2016). Land uses also significantly affect natural resources, and human interferences have had a considerable impact on the earth's surface in the last decades. Land uses are activities manipulating a specific type of land cover to produce, change, or conserve it (Kundu et al. 2017; Ghorbani et al. 2018).

According to the literature, numerous studies have recently been conducted on soil pollution to investigate toxic elements, especially in the developed countries. It has been reported that areas near industrial activities are considerably influenced by

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air, soil, and water pollution (Karimi et al. 2011; Naimi and Ayoubi 2013). Krishna and Govil (2008) in India, Alipour and Malekian (2016) and Nazarpour et al. (2017) in Iran, and Tripathee et al. (2016) in Nepal have examined metal concentrations, the spatial distribution of elements, and heavy metal concentration risk in the soil surrounding oil refineries.

As a review of the related literature shows, metal contamination mainly originates from anthropogenic activities. The oil industry activities have caused ecosystem instability and decreased biodiversity affecting the general environment of these areas (Kamalu and Wokocha 2011). High concentrations of heavy metals and complex organic compounds, including polycyclic and inorganic aromatic hydrocarbons, can be detected in crude oil. The concentration of these components depends on the type of oil exploited in different regions. Being toxic due to characteristics such as environmental non-degradability, excessive toxicity, accumulation, and carcinogenicity, heavy metals are among the ecological concerns of today's world and pollute terrestrial ecosystems (Cram et al. 2004; Micó et al. 2006; Fatoba et al. 2015). Having a long biological half-life, heavy metals are non-biodegradable and sustainable and can enter the human body through air, food, and water. Despite the fact that some heavy metals such as Mn, Fe, and Zn are essential and play a functional and structural role in biological systems (e.g., Fe in hemoglobin), others such as Cr are non-essential toxic metals. Some heavy metals such as Ni, Cu, and Zn also act as micronutrients at low concentrations but are toxic if taken at high concentrations (Rezaei Raja et al. 2016; Sobhanardakani 2017, 2019; Mohammadi et al. 2018; Davodpour et al. 2019). Exposure to high doses of heavy metals can cause adverse effects on human health. For example, exposure to Cd causes lung adenocarcinoma, lung cancer, and bone loss. Lead exposure can also disrupt the biosynthesis of hemoglobin, lead to a disorder of the central nerves system and hematopoietic system, and cause kidney damage, reduced learning ability, cancer, increased blood pressure, increased risk of Alzheimer's, and behavioral disorders in children (Sobhanardakani et al. 2018; Taati et al. 2020).

Oil contamination is among the main ecological challenges in oil-rich countries around the world. As the fourth-largest crude oil producer, Iran is also exposed to oil pollution during oil production and transportation, as most Iranian oil refineries are located in agricultural and urban areas (Soleimani et al. 2013).

Different indicators, including the I_{geo} index, ecological risk factor (E_i), integrated pollution index (IPI), pollution load index (PLI), Nemrow integrated pollution index (NIPI), and enrichment factor (EF) have been used for the ecological risk assessment of heavy metals in the surface soils. Models such as the potential ecological risk index (PERI) can also be employed for the assessment of relative poisonousness and the level of contamination with such elements in soils (Muller 1969; Caeiro et al. 2005; Liu et al. 2005; Yang et al. 2011; Zajusz-Zubek et al. 2015; Sabet Aghlidi et al. 2020).

Few types of research about soil contamination analyzing the toxic elements have been performed in the developing countries notably Iran. Arak is one of the most important industrial centers of Iran, which is host to numerous industries such as an oil refinery, a petrochemical corporation, a thermal power plant, aluminum manufacturing, and other factories, along with lead and zinc mines. On the other hand, Arak is considered to be one of the most important agricultural production centers of the country, and therefore, high levels of heavy metals in the soil can have an impact on human health and other living organisms. Therefore, this study was conducted in one of the major industrial areas in the southwest Arak, the Shazand Oil Refinery Complex, to investigate the environmental pollutants in the soil surrounding the refinery. Until now, no comprehensive and extensive research has been carried out on the potential ecological risks to the soils of the study area with regard to different land uses. The main objectives of the study were to investigate the accumulation and spatial distribution of heavy metals, namely Fe, Cd, Cr, Cu, Pb, Ni, Mn, and Zn (mg/kg) in the surface soils to assess their ecological risks and to identify their sources in various land uses (rangeland areas, agricultural areas, and industrial areas) around Shazand oil refinery.

Material and methods

Study area

Shazand Oil Refinery Complex (34.0048° N, 49.4746 °E) is located 17 km from Arak (capital of Markazi Province, Iran) at an altitude of 1900 m above sea level with a cold semi-arid climate. The average annual rainfall in the study area is 420 mm with a mean temperature of 12.1 °C. Petrographically and stratigraphically, visible protrusions including igneous and sedimentary formations are visibly observed in the study area (Gadimi et al. 2019; Taati et al. 2020). Different types of human impacts including agricultural, industrial (Shazand Oil Refinery Complex, Shazand Petrochemical Company, and Shazand Thermal Power Plant), and residential areas exist in the study area. Fig. 1 displays the sampling sites in the study area.

Sample preparation for analysis

Soil samples were collected in March 2020 from sampling points selected at different land uses from Oil Refinery and its surroundings. The samples were taken from 0–20-cm plots of soil after cleaning the organic materials on the surface. The sample's weight varied from 1 to 2 kg. Totally, 105 surface soil samples were collected from 21 sites. Of this, 40 samples were gathered from the rangeland area, 25 from the industrial area, and 40 from the agricultural area. The sampling stations were randomly selected. Five samples were collected from every station with a distance of 10 m from each other.

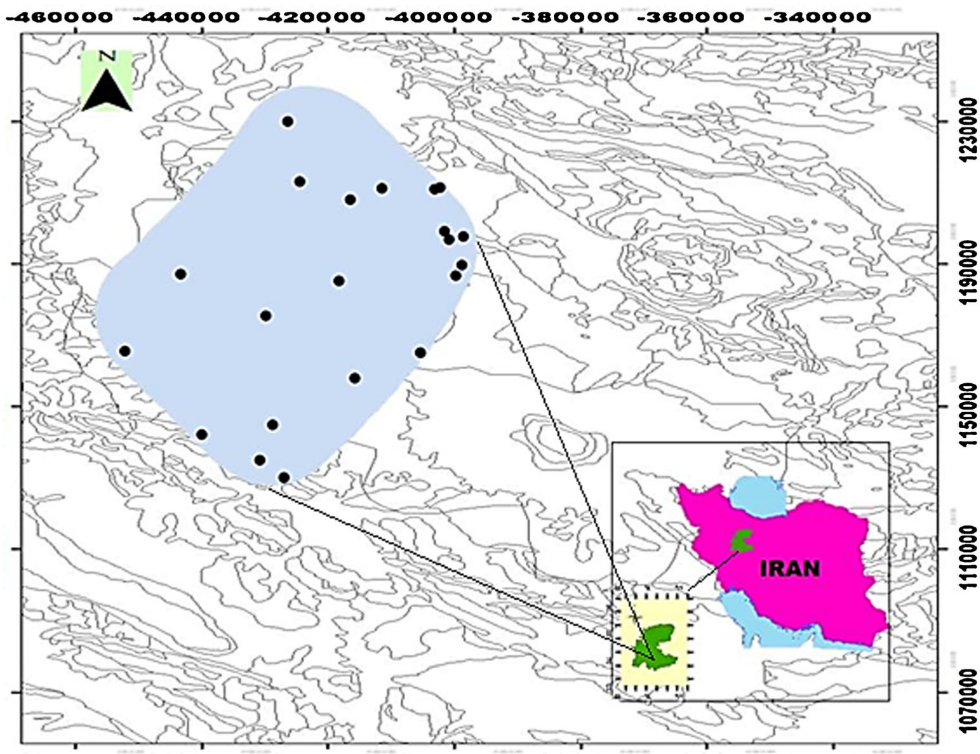

Fig. 1 Location of sampling sites from the study area

Soil analyses and quality control

The surface soil samples were dried at room temperature (25 °C). Each surface soil sample was then crushed and sieved by a 2-mm sieve to remove large debris. Heavy metals (Fe, Cu, Cd, Zn, Pb, Cr, Mn, and Ni) were determined by the diethylenetriamine penta-acetic acid (DTPA) method (Lindsay and Norvell 1978; Hosseinpur and Motaghian 2015). To this end, 20 mL DTPA extraction solution was added to 10 g of soil. After shaking for 120 min at a speed of 145 rpm, the samples were filtered by passing them through the Whatman® 42 paper. The volume of heavy metals in the extracted solution was calculated using the flame atomic absorption spectrometry (AAS, FS 240, Agilent, USA) (Chen et al. 2019). The pH and electrical conductivity were respectively measured by a pH meter (Rhoades 1996) and a conductivity meter (Thomas 1996; Hosseinpur and Motaghian 2015). The organic matter (OM) was determined by titration (Walkley and Black 1934). For each heavy metal, a recovery study was carried out to recheck its concentration more precisely with a higher accuracy and reliability. The performance indicators including the linear dynamic range (LDRs), limit of detection (LODs), relative standard deviation (RSD), and limit of quantification (LOQ) were determined for the heavy metals. The results are presented in Table 1. All soil samples were analyzed in Lorestan University Laboratory, Iran.

Risk assessment

Geo-accumulation index (I_{geo})

The geo-accumulation index (I_{geo}) was first presented by Muller in 1969 to assess the environmental risk (Muller 1969) using Eq. (1):

$$I_{geo} = \log2\left(\frac{C_n}{1.5\ B_n}\right) \quad (1)$$

where C_n represents the measured concentration (mg/kg) of the heavy metal in the soil samples and B_n shows the geochemical background value of the metal in the soil (Cai et al. 2015). This is a 7-degree index in the range of 5 < I_{geo} < 0, interpreted as reported in Table 2 (Nicholson et al. 2003; Benhaddya and Hadjel 2014a).

Potential ecological risk index (PERI)

The risk index (RI) is used to evaluate the degree of heavy metal pollution in soil (Kowalska et al. 2016), indicating the poisonousness of heavy metals and the environmental reactions (Benhaddya and Hadjel 2014a). RI is calculated using Eqs. (2) to (4):

$$PERI = \sum \quad {}^{n}_{i} = 1^{EI} \quad (2)$$

$$Ei = Ti \times Fi \quad (3)$$

Table 1 Performance indicator values for heavy metals

Experimental index	Zn	Pb	Mn	Cu	Fe	Cr	Ni	Cd
LDRs	0.0076–5.0	0.0098–6.0	0.0043–7.0	0.0032–4.0	0.0087–6.0	0.0054–6.0	0.0065–9.0	0.0043–6.0
LODs	0.00065	0.00054	0.00092	0.00011	0.00032	0.00021	0.00043	0.00019
RSDs	0.98	0.65	0.367	0.356	0.476	0.576	0.756	0.23
LOQ	0.0076	0.0098	0.0043	0.0032	0.0087	0.0054	0.0065	0.0043
Line equation	Y = 123.92x–3.09	Y = 12.46576–0.987	Y = 11.877–0.877	Y = 18.99–4.09	Y = 28.445–0.455	Y = 31.615–0.754	Y = 31.615–0.754	Y = 15.766–4.877
R^2	> 0.9998	> 0.9999	> 9996	> 0.9995	> 0.9994	> 0.9997	> 0.9994	> 0.9993

$$Fi = \frac{Ci}{Bi} \quad (4)$$

where E_i represents the potential ecological risk factor in the studied site and T_i is the toxic response factor of element i (5, 5, 5, 30, 1, 1, and 2 for Cu, Ni, Pb, Cd, Mn, Zn, and Cr, respectively) (Qingjie et al. 2008; Sobhanardakani 2018). F_i stands for the metal contamination factor in the soil samples, C_i shows the concentration of heavy metals, and Bi is the background value of the elements. The E_i and RI classifications (Benhaddya and Hadjel 2014a) are interpreted as reported in Table 2. As suggested by Caeiro et al. (2005), Liu et al. (2005), and Abrahim and Parker (2008), to evaluate the pollution level of heavy metals, the single pollution index (PI) for each heavy metal in land uses, the integrated pollution index (IPI), and the pollution load index (PLI) for each of the 8 elements in this study were calculated in accordance with Eqs. (5) and (7):

$$\mathrm{PI} = \frac{C}{S} \quad (5)$$

$$PLI = (PI1 \times PI2 \times PI3 \times \ldots \times PIn)1/n \quad (6)$$

$$IPI = mean\ (PIi) \quad (7)$$

where C (mg/kg) is the measured content of each element in the studied site, S (mg/kg) stands for the reference value of the elements, and n represents the number of examined elements. The PI, PLI, and IPI classifications are interpreted as reported in Table 2.

Nemrow integrated pollution index

The Nemrow index is extensively used as an effective method for estimating soil contamination by heavy metals. The main advantage of NIPI over other indicators is the possibility to assess the risk of soil contamination by all metals in the study area. NIPI is calculated from Eq. (8):

$$\mathrm{NIPI} = \sqrt{\frac{\mathrm{PI}^2_{\mathrm{max}} + \mathrm{pI}^2_{\mathrm{ave}}}{2}} \quad (8)$$

where $\mathrm{PI}^2_{\mathrm{max}}$ and $\mathrm{pI}^2_{\mathrm{ave}}$ respectively represent the maximum and average values of each pollutant index. The NIPI classifications are interpreted as reported in Table 3 (Yang et al. 2011).

Enrichment factor

The enrichment factor (EF) is calculated to assess the degree of pollution by heavy metals. This factor is calculated using Eq. (9) (Mandeng et al. 2019):

$$\mathrm{EF} = \frac{\left(\frac{c_i}{c_{\mathrm{ref}}}\right)_{sample}}{\left(\frac{c_i}{c_{\mathrm{ref}}}\right)_{\mathrm{background}}} \quad (9)$$

Table 2 The classification of I_{geo}, E_i, RI, PLI, IPI, PI, and IPI in relation to pollution and potential ecological risk

I_{geo} value	Pollution level	E_i value	Pollution level
$I_{geo} \leq 0$	Unpolluted	$E_i < 40$	Low
$0 < I_{geo} < 1$	Indicates unpolluted to moderately polluted	$40 < E_i < 80$	Moderate
$1 < I_{geo} < 2$	Moderately polluted	$80 < E_i < 160$	Appreciable
$2 < I_{geo} < 3$	Moderately to strongly polluted degrees	$160 < E_i < 320$	High
$4 < I_{geo} < 5$	Strongly to very strongly polluted	$E_i > 320$	Serious
RI values	Pollution level	PLI value	Pollution level
RI < 150	Low	PLI < 1	Unpolluted
150 < RI < 300	Moderate	1 < PLI < 2	Moderately polluted
300 < RI < 600	High	2 < PLI < 3	Strongly polluted
RI > 600	Significantly highest	PLI > 3	Extremely polluted
PI value	Pollution level	IPI value	Pollution level
PI ≤ 1	Low pollution	IPI < 1	Low
1 < PI ≤ 3	Moderate pollution	1 < IPI < 2	Middle
PI > 3	High pollution	IPI > 2	High

where C_i is the concentration of the target element in the studied site and C_{ref} represents the concentration of the reference element. Aluminum, magnesium, or iron are typically used in most studies as reference elements since they are the main constituents of the earth's crust (Zajusz-Zubek et al. 2015; Barbieri 2016). Magnesium was used in this study as the reference element. The descriptive classification of EF is interpreted as reported in Table 4 (Mandeng et al. 2019).

Statistical analyses

The experimental data were analyzed using SPSS V. 25 for Windows. The heavy metals standard deviation (SD) was calculated for each sampling site. The Kolmogorov-Smirnov (K-S) test was performed to test the normality of the distribution of the experimental data. The Pearson correlation coefficient (PCC), principal component analyses (PCA), and cluster analysis (CA) were run to identify different groups of metals. Moreover, the contour map of elements was generated by the kriging interpolation technique with the help of Arc GIS V. 10.3.

Table 3 Classification of NIPI value

NIPI value	Pollution level
NIPI < 0.7	Non-pollution
0.7 < NIPI < 1	Warning line of pollution
1< NIPI < 2	Low level of pollution
2 < NIPI < 3	Moderate level of pollution
NIPI > 3	High level of pollution

Results and discussion

Characteristics of the heavy metals in the soil samples

Table 5 summarizes the statistical data on the heavy metals and some chemical properties (pH, EC, and OM) of the analyzed surface soil samples along with the soil background values. As shown in the table, the concentrations of Fe, Cu, Ni, Cd, Pb, Cr, Mn, and Zn respectively varied from 0.776 to 33.6, 0.284 to 2.06, 0.074 to 3.73, 0.001 to 0.698, 0.010 to 104, 0.004 to 0.092, 1.11 to 154, and from 0.306 to 41.8 mg/kg with average concentrations of 7.98, 1.27, 1.20, 0.042, 3.84, 0.025, 44.3, and 3.57 mg/kg, and median concentrations of 5.77, 1.26, 0.644, 0.020, 1.82, 0.024, 15.3, and 1.83 mg/kg, respectively. As can be seen, the average concentrations of all the heavy metals were less than their background values. The background values for Fe, Zn, Pb, Cd, Cr, Cu, Mn, and Ni were respectively 47200, 48.5, 36.5, 0.300, 20.8, 28.3, 850, and 41.6 (Azimzadeh and Khademi 2013; Mohammadi et al. 2015; Sobhanardakani et al. 2016). The coefficient of variation (CV) is the most widely used factor

Table 4 Classification of the EF value

EF value	Pollution level
EF < 1	No enrichment
1 < EF < 3	Minor enrichment
3 < EF < 5	Moderate enrichment
5 < EF < 10	Moderately severe enrichment
10 < EF < 25	Severe enrichment
25 < EF < 50	Very severe enrichment
EF > 50	Extremely severe enrichment

Table 5 Descriptive statistics of soil's heavy metal in the rangeland, industrial, and agricultural area (mg /kg)

Heavy metal	Area	Minimum	Maximum	Mean	Median	SD	CV %	K - S_p
Fe	All	0.776	33.620	7.976	5.774	6.042	75.8	0.000
	Rangeland area	0.776	16.964	5.642	3.982	3.768	66.8	0.000
	Industrial area	1.556	19.926	7.292	6.982	4.565	62.6	0.107
	Agricultural area	2.242	33.620	10.737	10.585	7.525	70.1	0.007
Cu	All	0.284	2.060	1.272	1.264	0.394	31	0.200
	Rangeland area	0.284	1.948	1.187	1.176	0.391	33	0.200
	Industrial area	0.308	1.896	1.146	1.078	0.367	32.1	0.200
	Agricultural area	0.552	2.060	1.437	1.509	0.364	25.3	0.005
Ni	All	0.074	3.734	1.199	0.644	0.995	83	0.000
	Rangeland area	0.074	2.972	0.770	0.403	0.795	103.2	0.000
	Industrial area	0.108	2.806	1.150	1.294	0.842	73.2	0.012
	Agricultural area	0.164	3.734	1.657	1.675	1.079	65.1	0.003
Cd	All	0.001	0.698	0.042	0.020	0.090	214.5	0.000
	Rangeland area	0.001	0.456	0.044	0.024	0.075	169.3	0.000
	Industrial area	0.001	0.698	0.041	0.014	0.137	338.2	0.000
	Agricultural area	0.002	0.346	0.041	0.024	0.068	164.7	0.000
Pb	All	0.010	103.700	3.841	1.820	11.281	293.7	0.000
	Rangeland area	0.010	103.700	5.422	1.370	16.442	303.3	0.000
	Industrial area	0.010	3.840	1.458	1.120	1.086	74.5	0.129
	Agricultural area	0.540	51.760	3.750	2.870	7.881	210.2	0.000
Cr	All	0.004	0.092	0.025	0.024	0.014	55.5	0.000
	Rangeland area	0.004	0.040	0.024	0.024	0.010	40.6	0.022
	Industrial area	0.006	0.092	0.028	0.022	0.020	69.9	0.037
	Agricultural area	0.004	0.076	0.024	0.022	0.013	53.9	0.048
Mn	All	1.107	153.901	44.295	15.331	42.510	96	0.000
	Rangeland area	3.971	153.901	31.226	8.882	38.636	123.7	0.000
	Industrial area	4.319	147.805	50.409	40.648	45.346	90	0.000
	Agricultural area	1.107	118.504	53.544	58.291	42.117	78.7	0.000
Zn	All	0.306	41.827	3.567	1.826	5.386	151	0.000
	Rangeland area	0.702	26.608	4.590	2.321	5.601	122	0.000
	Industrial area	0.306	5.809	1.679	1.380	1.142	68.1	0.001
	Agricultural area	0.774	41.827	3.725	2.134	6.465	173.6	0.000
PH	All	5.540	8.390	7.909	8.170	0.689	8.7	0.000
	Rangeland area	5.540	8.390	7.631	8.115	1.012	13.3	0.000
	Industrial area	7.560	8.350	8.047	8.170	0.278	3.5	0.001
	Agricultural area	7.440	8.390	8.101	8.185	0.253	3.1	0.004
EC	All	113.900	318.000	177.330	157.900	48.752	27.5	0.000
	Rangeland area	113.900	289.000	159.316	145.750	40.917	25.7	0.000
	Industrial area	146.800	275.000	194.480	168.000	44.301	22.8	0.000
	Agricultural area	133.600	318.000	184.625	159.350	53.666	29.1	0.000
OM	All	0.001	5.840	1.042	0.806	1.180	113.2	0.000
	Rangeland area	0.001	5.840	0.863	0.644	1.342	155.5	0.000
	Industrial area	0.001	5.310	1.192	0.941	1.244	104.4	0.024
	Agricultural area	0.001	3.700	1.129	1.142	0.951	84.3	0.170

SD, standard deviation; *CV*, coefficient of variation

for explaining variability (Yongming et al. 2006; Zhang et al. 2007). As indicated, Cu reflected the lowest CV (31.0%) followed by Cr (55.5%), and Pb, Cd, and Zn had CVs of more than 90% (294, 215, and 151%, respectively). The pH values

Table 6 The correlation matrix between the elements in soil specimens

Heavy metal	Fe	Cu	Ni	Cd	Pb	Cr	Mn	Zn
All								
Fe	1							
Cu	0.454**	1						
Ni	0.769**	0.523**	1					
Cd	0.138	0.153	0.128	1				
Pb	0.119	0.083	0.004	0.587**	1			
Cr	0.098	− 0.006	0.118	0.100	− 0.050	1		
Mn	0.636**	0.368**	0.691**	0.210*	0.226*	0.214*	1	
Zn	0.038	0.021	− 0.084	0.501**	0.692**	− 0.046	0.006	1
Rangeland area								
Fe	1							
Cu	0.778**	1						
Ni	0.713**	0.652**	1					
Cd	0.540**	0.320*	0.140	1				
Pb	0.461**	0.240	0.120	0.961**	1			
Cr	0.087	0.329*	− 0.062	0.142	0.035	1		
Mn	0.743**	0.516**	0.569**	0.519**	0.532**	0.045	1	
Zn	0.478**	0.261	0.042	0.745**	0.627**	0.231	0.297	1
Industrial area								
Fe	1							
Cu	0.612**	1						
Ni	0.778**	0.534**	1					
Cd	0.182	0.253	0.254	1				
Pb	0.693**	0.379	0.613**	0.137	1			
Cr	− 0.125	− 0.536**	− 0.066	− 0.002	0.152	1		
Mn	0.742**	0.407*	0.815**	0.141	0.525**	0.091	1	
Zn	0.085	0.350	0.240	0.117	0.634**	− 0.019	0.043	1
Agricultural area								
Fe	1							
Cu	0.137	1						
Ni	0.739**	0.313*	1					
Cd	− 0.028	− 0.098	0.078	1				
Pb	− 0.103	− 0.240	− 0.132	0.744**	1			
Cr	0.281	0.303	0.402*	0.272	− 0.211	1		
Mn	0.563**	0.147	0.692**	0.052	− 0.102	0.463**	1	
Zn	− 0.121	− 0.269	− 0.175	0.739**	0.968**	− 0.193	− 0.156	1

**$p < 0.01$, *$p < 0.0$

showed no significant changes in different land uses. The pH values of the soil in the study area fell within a range of neutral to alkaline pH.

Correlation coefficient analysis

The inter-element relationships can reveal interesting facts about heavy metal sources (Rodriguez et al. 2008). Table 6 shows the Pearson correlation coefficients (PCCs) for the 8 selected heavy metals in the soil samples. In the present study, the PCC was used as an indicator of the relationship between different heavy metals. As the data in the table indicate, at $p < 0.01$, there was a significant direct correlation between the heavy metal pairs of Fe-Ni (0.769) and Pb-Zn (0.692), suggesting the same origin of these elements. At $p < 0.01$, a strong positive correlation was observed between Cd and Pb (0.961) in the rangeland area. A direct correlation was also detected between the Ni-Mn pair (0.815) in the industrial area. Finally, there was a direct correlation between the Pb-Zn pair (0.968) in the agricultural area.

Table 7 Variance description and component models of the element

Heavy metal	All			Rangeland area			Industrial area			Agricultural area		
	PC1	PC2	PC3	PC1	PC2	PC3	PC1	PC2	PC3	PC1	PC2	PC3
Fe	0.870	0.061	0.055	0.860	0.396	0.067	0.900	0.131	0.209	− 0.065	0.889	− 0.015
Cu	0.702	0.057	− 0.205	0.829	0.117	0.385	0.466	0.308	0.741	− 0.188	0.043	0.774
Ni	0.916	− 0.049	0.084	0.917	− 0.068	− 0.109	0.886	0.213	0.137	− 0.032	0.879	0.253
Cd	0.149	0.790	0.149	0.208	0.955	0.048	0.216	0.194	0.170	0.915	0.052	0.218
Pb	0.079	0.897	− 0.053	0.174	0.933	− 0.088	0.602	0.716	− 0.110	0.940	− 0.066	− 0.221
Cr	0.062	− 0.006	0.962	0.027	0.082	0.961	0.075	0.085	− 0.941	0.058	0.356	0.765
Mn	0.794	0.138	0.266	0.728	0.423	− 0.101	0.930	0.008	− 0.045	− 0.014	0.826	0.208
Zn	− 0.070	0.866	− 0.086	0.092	0.818	0.253	− 0.019	0.972	0.088	0.939	− 0.105	− 0.223
Initial eigenvalue	2.757	2.208	1.081	2.880	2.813	1.174	3.093	1.660	1.547	2.648	2.391	1.437
% of variance	34.458	27.602	13.511	35.995	35.158	14.669	38.662	20.750	19.333	33.101	29.889	17.966
Cumulative %	34.458	62.060	75.571	35.995	71.154	85.822	38.662	59.412	78.745	33.101	62.990	80.956

Element source identification

Based on the results of PCA, the analyzed element contents are grouped into a three-component model (PC1, PC2, and PC3). Table 7 lists the PCA results of the studied heavy metals in the rangeland, agricultural, and industrial areas. As illustrated in the table, PC1 described 34.5% of the total data variability showing maximum positive loadings for Fe (0.870), Ni (0.916), Cu (0.702), and Mn (0.794). However, it reflected moderate and low positive loadings for Cd (0.149), Pb (0.079), and Cr (0.062) respectively. The PC2 accounted for 27.6% of the total data variability showing maximum positive loadings for Cd (0.790), Pb (0.897), and Zn (0.866). The PC3 describes 13.5% of the total data variability showing maximum positive load for Cr (0.962). The PC1 in the rangeland area described 36.0% of the total variability showing maximum positive loads for Fe (0.860), Ni (0.917), Cu (0.828), and Mn (0.728). The PC2 explained 35.2% of the total data variability showing maximum positive loadings for Cd (0.955), Pb (0.933), and Zn (0.818). The PC3 accounted for 14.7% of the total variability showing maximum positive loading for Cr (0.961). The PC1 in the industrial area explained 38.7% of the total data variability reflecting maximum positive loadings for Mn (0.972), Fe (0.900), and Ni (0.886). The PC2 described 20.8% of the total variability showing maximum positive loadings for Pb (0.716) and Zn (0.972). The PC3 explained 19.3% of the total data variability reflecting the maximum positive loading for Cu (0.741) and the highest negative variation loading for Cr (0.941). The PC1 in the agriculture area described 33.1% of the total data variability showing maximum positive loadings for Cd (0.915), Pb (0.940), and Zn (0.939). The PC2 explained 29.9% of the total data variability reflecting maximum positive loadings for Fe (0.889), Ni (0.879), and Mn (0.826). The PC3 accounted for 18.0% of the total variability showing maximum positive loadings for Cr (0.765) and Cu (0.774).

Cluster analysis

Cluster analysis is a multivariate technique for analyzing correlation coefficients to obtain similarity coefficients and plot a dendrogram. The cluster tree connects the subjects of the same weight to create larger clusters and assess similarities between specimens (Anazawa et al. 2004; Nguyen et al. 2005). Based on the dendrogram (Fig. 2), in the industrial and agriculture areas, elements were divided into 3 clusters. This was consistent with the results obtained from the PCA analyses where Cu, Ni, Cd, Pb, Cr, and Cu were in the same cluster. However, Fe and Mn fell into separate groups. Similarly, the elements in the rangeland area were divided into 3 clusters, and Cu, Fe, Ni, Cr, Cd, and Zn fell in the same cluster. Despite a negligible difference between these metals, Pb and Mn fell into separate groups suggesting different origins of these elements. The elements in the industrial and agricultural areas were divided into 3 clusters, and Cu, Cd, Ni, Pb, Cr, and Zn fell in the same cluster, whereas Fe and Mn fell in separate groups.

Geo-accumulation index (I_{geo})

As shown in Table 8, based on the results, the observed mean I_{geo} values could be classified as "unpolluted" for all elements. The maximum I_{geo} values for Fe, Cu, Ni, Cr, Mn, and Zn (− 11.0, − 7.22, − 9.72, − 8.41, − 10.2, and − 0.798) were in the unpolluted range, respectively. However, the maximum I_{geo} values for Cd and Pb (0.633 and 0.921) were in the unpolluted to the moderately polluted range. The maximum I_{geo} values for Cd and Pb were observed in the industrial and rangeland areas.

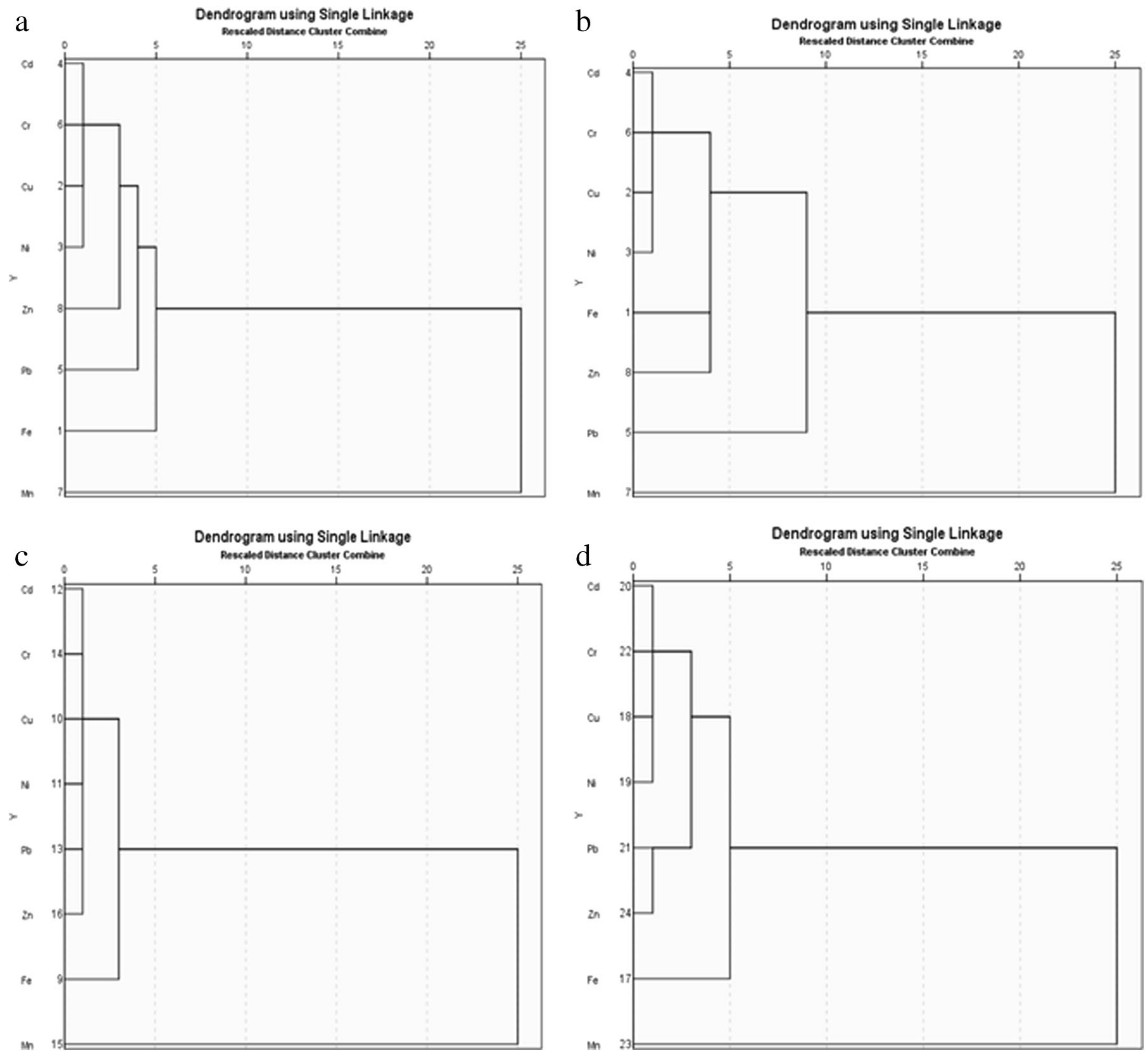

Fig. 2 Loading plot of analyzed elements in the space described by three principal components (PC1, PC2, and PC3) All (**a**), rangeland area (**b**), industrial area (**c**), Agricultural area (**d**)

Potential ecological risk index (PERI)

As seen in Table 9, based on the results obtained, the observed mean E_i values could be categorized as "low ecological risk" for all elements. The maximum E_i value of 69.8 for Cd indicated that it could pose "moderate ecological risk," a result consistent with that obtained for the I_{geo} index. Table 11 shows the PERI for the total metal concentration in soil

Table 8 I_{geo} values for elements in soil samples

Elements	I_{geo} value			Soil quality
	Mean	Minimum	Maximum	
Fe	− 13.4725	− 16.4773	− 11.0402	Unpolluted
Cu	− 5.14357	− 4.36505	− 7.22373	Unpolluted
Ni	− 6.31572	− 4.06275	− 9.71981	Unpolluted
Cd	− 4.631	− 8.81378	0.633302	Unpolluted to moderately polluted
Pb	− 5.11167	− 12.4186	0.921485	Unpolluted to moderately polluted
Cr	− 10.5264	− 12.9293	− 8.4057	Unpolluted
Mn	− 5.81474	− 3.05042	− 10.1701	Unpolluted
Zn	− 5.00612	− 7.89327	− 0.7985	Unpolluted

Table 9 E_i values for elements in soil samples

Elements	E_i value			Ecological risk factor of element
	Mean	Minimum	Maximum	
Cu	0.225	0.050	0.364	Low
Ni	0.144	0.009	0.449	Low
Cd	4.212	0.100	69.800	Moderate
Pb	0.526	0.001	14.205	Low
Cr	0.002	0.000	0.009	Low
Mn	0.052	0.001	0.181	Low
Zn	0.074	0.006	0.862	Low

specimens. Based on the ecological risk index value, the whole research area could be categorized as being at "low ecological risk."

As shown in Table 10, based on the results obtained, the observed mean PI values for all the elements indicated that they could cause "low contamination." However, the maximum PI values of 2.33 and 2.84 for Cd and Pb respectively, which were consistent with the results obtained from the I_{geo} index and E_i factor, could be considered as causing "moderate contamination." The IPI and PLI indexes for total metal concentration in the soil samples are shown in Table 11. Based on the IPI value obtained, all the studied soils could be categorized as "low" polluted soils. As the PLI values show, the whole research scope could be categorized as "unpolluted."

Nemrow integrated pollution index and enrichment factor

As shown in Table 12, the NIPI values for all the heavy metals under study showed they could be classified as non-pollution. The NIPI value of 1.65 for Cd meant it could cause a low level of pollution while the corresponding value of 2.01 for Pb implied it would cause a moderate level of pollution. These results were consistent with those obtained from the PI index.

Table 10 PI of elements in soil samples

Elements	*N*	PI			Number of samples		
		Minimum	Maximum	Mean	Low	Middle	High
Fe	105	0.000	0.001	0.000	105	0	0
Cu	105	0.010	0.073	0.045	105	0	0
Ni	105	0.002	0.090	0.029	105	0	0
Cd	105	0.003	2.327	0.140	102	3	0
Pb	105	0.000	2.841	0.105	103	2	0
Cr	105	0.000	0.004	0.001	105	0	0
Mn	105	0.001	0.181	0.018	105	0	0
Zn	105	0.006	0.862	0.038	105	0	0

As seen in Table 12, based on the results, the EF values for Fe, Cu, Ni, and Cr could be classified as no enrichment while the EF values of 2.69, 2.02, and 1.41 for Cd, Pb, and Zn respectively could be classified as minor enrichment.

Descriptive statistics for the total heavy metals concentrations in the soil samples of the study area are summarized in Table 5. As shown in the table, the mean concentrations of Fe, Cu, Ni, Cd, Pb, Cr, Mn, and Zn (7.98, 1.27, 1.20, 0.042, 3.84, 0.025, 44.3, and 3.57 mg/kg, respectively) were lower than those reported as the background values. In this study, the average heavy metal concentrations measured in surface soil samples were in the order of Mn > Fe > Pb > Zn > Cu > Ni > Cd > Cr for the rangeland area, Mn > Fe > Zn > Pb > Cu > Ni > Cd > Cr for the industrial area, and Mn > Fe > Pb > Zn > Ni > Cu > Cd > Cr for the agricultural area. The high average values of heavy metals in the rangeland and agriculture areas can be attributed to the industrial activities and traffic volumes in those areas. In studying the soil samples of Tehran Refinery, Iran, Pourang and Noori (2014) reported Mn > Zn > Cr > V > Ni > Pb as the order of abundance of the elements. In their study on the soil samples around Tehran Refinery, Iran, Seilsepour and Bigdeli (2008) also reported Mn > Zn > Cr > Pb > Ni for the concentrations of the elements. Gharib and Al Sarawi (2018) reported the level of the elements in the soil samples surrounding three southern oil refineries in Kuwait as Mn > Zn > Ni > Cr > V > Pb > Cu > Fe > Cd. Benhaddya and Hadjel (2014b) studied soil samples around an oil field in southeastern Algeria and found the descending order of Mn > Zn > Pb > Cu > Ni for the average concentration of the metals. Table 13 compares the element levels (mg/kg)

Table 11 RI, IPI, and PLI of elements in soil samples

Index	*N*	Minimum	Maximum	Mean	Median	Std. Deviation
RI	105	0.320	70.756	5.235	2.826	10.119
IPI	105	0.008	0.629	0.056	0.036	0.080
PLI	105	0.002	0.060	0.014	0.012	0.009

Table 12 NIPI and Enrichment factor of elements in soil samples

	Fe	Cu	Ni	Cd	Pb	Cr	Mn	Zn
NIPI	0.00051	0.06049	0.06665	1.64819	2.01033	0.00323	0.13322	0.61203
EF	0.003243	0.86279	0.552877	2.694437	2.019432	0.022695	1	1.411388

in the soil samples of the present study with those reported in the literature. As can be seen, no significant difference was observed between the heavy metal concentrations in different land uses (rangeland, industrial, and agriculture areas). Nadal et al. (2007) reported that soil samples collected from various industrial and residential areas were contaminated under the activities of petrochemical industries; however, their results showed no significant difference between various collection areas for some metals (Nadal et al. 2007).

The coefficient of variation (CV) indicates the variability in the concentrations of soil heavy metals (Yongming et al. 2006; Zhang et al. 2007). In our study, the CV of heavy metals increased in order of Cu (31.0%) < Cr (55.5%) < Fe (75.8%) < Ni (83.0%) < Mn (96.0%) < Zn (151%) < Cd (214%) < Pb (294%). A CV below 20.0% is considered as low variability, between 21.0 to 50.0% and 51.0 to 100% is regarded as medium and high variability, respectively, and above 100% is regarded as exceptionally high variability (Taati et al. 2020). Based on the results, Cu showed the lowest CV (31.0%) followed by Cr (55.5%) while Pb, Cd, and Zn showed CVs more than 100% (294, 215, and 151%, respectively) suggesting a wide range of variability. The relatively low CVs of the

Table 13 Comparison of elements level (mg/kg) in the soil samples of the present study with the literature data

Study area	Element								Reference
	Fe	Cu	Ni	Mn	Pb	Cd	Zn	Cr	
Iran	7.976	1.272	1.199	44.295	3.841	0.042	3.567	0.025	Present study
India	-	154.4	48.9	-	41.8	-	128.2	221.7	Krishna and Govil (2008)
China	-	38.8	-	-	69.4	0.23	158.6	-	Li et al. (2009)
China	-	-	20.52	-	56.38	-	66.15	43.01	Cai et al. (2010)
Algeria	-	0.44	15.21	0.86	14.28	-	1.7	-	Benhaddya and Hadjel (2014b)
India	-	-	45.20	-	87.84	1.68	-	74.10	Reza et al. (2014)
Iran	-	-	43.60	665.06	35.08	-	130.36	87.15	Pourang and Noori (2014)
India	6932	13.52	18.78	-	12.52	-	-	8.29	Tiwari et al. (2011)
Egypt	-	-	35.78	-	130.97	-	98	167	El-Taher and Abdelhalim (2014)
USA	6169	-	-	368	-	-	-	-	Zhang et al. (2015)
Saudi Arabia (North)	-	-	-	-	-	-	73.33	103.62	El-Taher et al. (2016)
Azerbaijan	-	-	-	-	29.2	0.18	47.9	19.9	Khalilova and Mammadov (2016)
Nepal	-	-	17.31	-	21.2	0.12	66.86	38.83	Tripathee et al. (2016)
Iran (Abadan)	-	-	75.7	-	10	1.8	-	82.8	Alipour and Malekian (2016)
Iran (Tabriz)	-	-	76.3	-	5	2.6	-	52	Alipour and Malekian (2016)
Iran (Esfahan)	-	-	122	-	9	4.4	-	9	Alipour and Malekian (2016)
Iran (Shiraz)	-	-	77.4	-	13	2.2	-	13	Alipour and Malekian (2016)
Iran (Tehran)	-	-	46.5	-	6.1	1.1	-	6.1	Alipour and Malekian (2016)
Saudi Arabia	-	-	15.97	-	6.5	0.11	20.38	14.88	Hasayen et al. (2017)
Saudi Arabia	-	-	2.61	-	26.9	1.65	21.1	17.4	Al-Wabel et al. (2017)
Iran	-	75.80	94.06	-	251.20	0.69	132.84	141.48	Nazarpour et al. (2017)
Saudi Arabia	-	-	2.67	-	2.27	39.9	1.14	2.70	Alshahri and El-Taher (2018)
Kuwait	17.15	23.79	112.78	404.27	27.71	0.08	151.08	56.23	Gharib and Al Sarawi (2018)
Iraq	31.50	-	0.116	-	-	0.227	-	-	Khudhur (2018)
Iran	2.0	97	84	472	43	0.43	165	60	Mokhtarzadeh et al. (2020)

heavy metals have been related to natural resources, while the fairly high CVs of metals have been associated with man-made influences (Yongming et al. 2006; Zhang et al. 2007). Thus, high levels of Pb and Cd may be related to anthropogenic activities.

PCA has been used in various fields such as in analyzing water quality as well as determining the soil, sediment, and environmental pollution sources (Spencer 2002; Borůvka et al. 2005; Chen et al. 2007). PCA has also been used to explain the anthropogenic and geogenic sources of heavy metals (Mokhtarzadeh et al. 2020). In this study, the PCA was performed to find sources of soil contamination. Based on the results, the heavy metal contents were categorized into a three-component model accounting for 76.0% of the total variance. Furthermore, three main components were found for every land use indicating three different sources of elements in this area. Ghassemi Dehnavi et al. (2019) and Taati et al. (2020) also reported three main components for heavy metals in the surface soil of the Arak industrial area. In general, the principal component (PC1) presented high loadings for Fe, Cu, Ni, and Mn. The second principal component (PC2) showed loadings for Cd, Pb, and Zn. The third principal component (PC3) showed a high Cr loading. Based on the PCA results for the heavy metal contents in the rangeland area, the evaluated heavy metal contents were classified into a three-component model explaining 86.0% of the total variance. Ghassemi Dehnavi et al. (2019) also reported a high Ni concentration in the soil samples surrounding Kermanshah Refinery in Iran. In the rangeland area, the principal component (PC1) presented high Fe, Ni, Cu, and Mn loadings. The PC2 showed loadings for Cd, Pb, and Zn. The PC3 showed a high loading for Cr. Based on the PCA results for the heavy metal contents in the industrial region, the heavy metal contents evaluated in a three-component model accounted for 79.0% of the total variance. In the industrial area, the PC1 presented high loadings for Mn, Fe, and Ni. The PC2 reflected loadings for Pb and Zn. The PC3 indicated a high positive Cu loading and a high negative Cr loading. Based on the PCA of the elements in the agricultural area, the analyzed elements were classified into a three-component model explaining 81.0% of the total variance. In the agricultural area, the PC1 presented high loadings for Cd and Zn. The PC2 exhibited loadings for Fe, Ni, and Mn. The PC3 showed high positive loadings for Cu and Cr. The principal component of soil in the agricultural area showed a significant amount of Cd suggesting fertilizers as the main origin of this heavy metal. Based on the dendrogram, the elements in the study area were divided into 3 clusters. This was in good agreement with the results of the PCA analysis. As shown in Fig. 3, the Cd hotspots were mainly detected in the east and west parts of the study area and could be attributed to human activities, especially agricultural practices. The existence of other industries such as the petrochemical industry and the thermal power plant around the refinery also could cause soil contamination by heavy elements. Wang and Qin (2006) maintained that industrial activities and petroleum refineries are the major causes of heavy metal emissions and thus high soil contamination by heavy metals in those areas. Moreover, agricultural practices, especially the use of fertilizers, could also be considered an important source of chromium and copper (Mirzaei et al. 2014). The hotspots in the Cr distribution pattern were mainly detected in the west, northwest, and southwest parts of the study area. The hotspot Cu regions were mainly found in the south, southwest, and west parts of the study area. The Fe and Ni hotspots were mainly detected in the west of the study area. In the case of Mn distribution, most areas on the map showed high concentrations as shown in Fig. 3e. The hotspot Pb and Zn regions were mainly found in the east of the study area.

The I_{geo} values indicated that the soils in the research area are unpolluted to moderately polluted. Among the environmentally toxic heavy metals, Cd and Pb were found to be considerably accumulated in the studied soils as indicated by their maximum I_{geo} values of 0.633 and 0.921, respectively. In contrast, the I_{geo} value for Fe (− 13.5), Cu (− 5.14), Ni (− 6.32), Cr (− 10.5), Mn (− 5.81), and Zn (− 5.01) were less than zero suggesting that the soils are unpolluted by these heavy metals. Benhaddya and Hadjel (2014b) also reported negative I_{geo} indexes for Cu, Ni, and Mn based on their average rates in the industrial areas of Algeria (Hassi Messaoud), whereas the I_{geo} index for Pb placed it in the moderately polluted category. As reported by Alshahri and El-Taher (2018), the Cd level in the surface soil near an oil refinery in Saudi Arabia was classified as extremely polluted.

Based on the PI values, the trace elements (Fe (0.000), Cu (0.045), Ni (0.029), Cd (0.140), Pb (0.105), Cr (0.001), Mn (0.018), and Zn (0.038) were placed in the "low contamination" category. The PI values of Cd and Pb could be considered as "moderate contamination." Alshahri and El-Taher (2018) also reported the highest mean PI value for Cd.

As the EF values show, the heavy metals in the research area decreased in the order of Cd (2.69) > Pb (2.02) > Zn (1.41) > Mn (1.00) > Cu (0.863) > Ni (0.553) > Cr (0.023) > Fe (0.003). In this study Cd, Pb and Zn had EF values greater than 1.0; thus, they could be attributed to anthropogenic activities. Taati et al. (2020) also reported EF values as As > Cd > Pb > Zn > Ni > Cu in the surface soil of Arak industrial area. According to Mokhtarzadeh et al. (2020), the enrichment factors for Cd, Pb, and Zn were significant in the soil samples collected from the Middle East oil refinery zone.

Based on the PERI (E_i) of the individual elements (Cd, Cu, Ni, Pb, Cr, Mn, and Zn) and their classifications, every single metal showed a low ecological risk factor except for Cd that reflected a considerable ecological risk and could be classified as a moderate ecological risk factor in the study area. On the

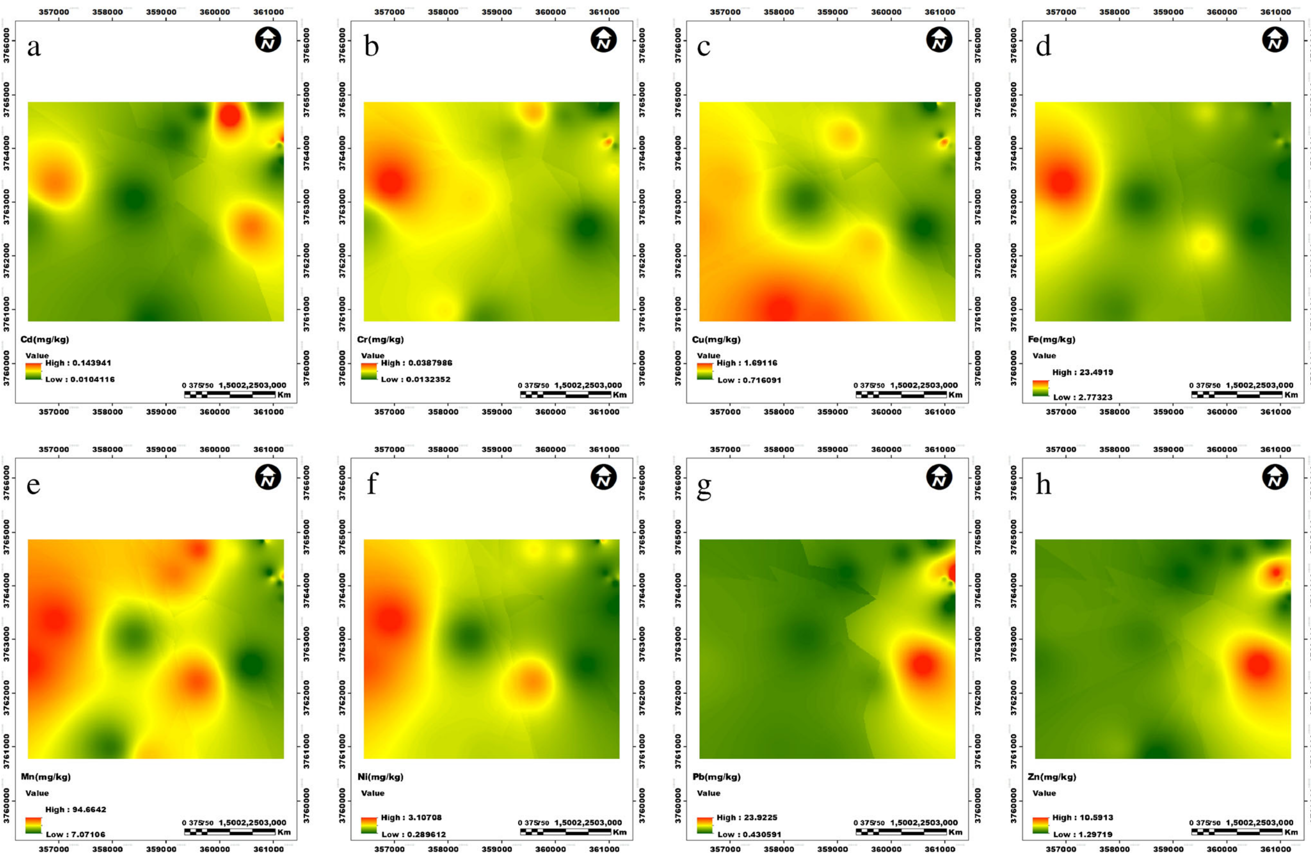

Fig. 3 Spatial distribution of Cd (**a**), Cr (**b**), Cu (**c**), Fe (**d**), Mn (**e**), Ni (**f**), Pb (**g**), and Zn (**h**) content in soil specimens around of Shazand refinery

whole, the PERI (E_i) values in the research area decreased in the order of Cd (4.21) > Pb (0.526) > Cu (0.225) > Ni (0.144) > Zn (0.074) > Mn (0.052) > Cr (0.002).

As the PI values show, the heavy metals in the study area decreased in the order of Cd (0.140) > Pb (0.105) > Cu (0.045) > Zn (0.038) > Mn (0.018) > Cr (0.001) > Fe (0.000). The maximum PI values of 2.33 and 2.84 for Cd and Pb respectively indicated that they could cause moderate contamination. The IPI value (0.056) for all the studied soils could be regarded as low polluted soils. Based on the PLI values (0.014), the whole research area could be categorized as unpolluted.

The calculated values for the PI, IPI, PLI, EF, and NIPI indices showed that considering the site class, the study area is somewhat influenced by industrial, agricultural, and transportation activities. The surface soils of the study area are mainly influenced by Cd and Pb contamination originated from human activities. Overall, the EF, PERI, and NIPI values confirmed the results obtained from calculating the I_{geo} index.

Conclusions

This study investigated and evaluated surface soil contamination by Fe, Cd, Cr, Cu, Pb, Ni, Mn, and Zn of the lands around Shazand Oil Refinery in Iran. The results showed that the rates of all the heavy metals are lower than their background values. However, the I_{geo} index showed that Cd and Pb with unpolluted to moderately polluted index values of 0.633 and 0.921 respectively exist in the soil. However, the I_{geo} index for other elements showed that the study area is not contaminated by those heavy metals. The average E_i value of 69.8 for Cd indicated that its ecological risk factor could be categorized as moderate in some samples. The average PI values suggested that 2.80% and 1.90% of the surface soil samples are highly polluted by Cd and Pb. The NIPI value indicated low and moderate contamination index values of 1.65 and 2.01 respectively for Cd and Pb in the soil samples. Based on the EF values of 2.69, 2.02, and 1.41 for Cd, Pb, and Zn respectively, the enrichment factor of these elements could be classified as minor. However, the RI value indicated that the whole research area could be classified as a “low ecological risk.” The IPI values for all the soil samples in the study area were categorized as “low contaminated soil.” The pollution load index (PLI) showed that the whole study area could be categorized as unpolluted. Based on HCA, PCA, and spatial distribution pattern of the elements, human activities are the main source of soil contamination and along with geogenic features which play significant roles in soil contamination by heavy metals in the study area. Thus, as it is likely that heavy metals will increase in the soil in the future, it is suggested that further studies be conducted to determine the effect of heavy metal bioavailability.

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Data availability Due to the nature of this research, participants of this study did not agree for their data to be shared publicly, so supporting data is not available.

Declarations

Ethical approval and consent to participate This article does not contain any studies with animals and human subjects. The authors confirm that all the research meets ethical guidelines and adheres to the legal requirements of the study country.

Consent for publication The authors declare that this manuscript does not contain any individual person's data and material in any form.

Conflict of interest The authors declare no competing interests.

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