Geochemical determination and pollution assessment of heavy metals in agricultural soils of south western of Iran

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Abstract

Soil contamination with heavy metals due to the application of fertilizers and biocides in agricultural activities is a potential threat for human health through the food chain. The present work was designed to study the spatial distribution of heavy metals, pollution level and possible reasons for their contamination in agricultural soils of Aghili plain, Khuzestan, Iran. The median concentrations of As, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, V, Zn, and Hg were 2.90, 0.29, 8.10, 39.0, 17.75, 354.0, 0.97, 58.35, 5.90, 34.0, 42.0, and 0.01 mg/kg, respectively. The results revealed that average concentrations of all studied heavy metals with an exception of Co, Cu, and Ni, were lower than background values. Analysis of source identification showed that Zn, Pb, and Cu ($P < 0.01$, $r > 0.9$) and Co, Cr, Mn, Ni, and V ($P < 0.01$, $r > 0.7$) were mainly from anthropogenic. In addition, Cd probably was originated from agricultural activities (application of manure and phosphorous fertilizers). Enrichment factor values of all metals (except Ni), were in the range of non to moderate enrichment ($EF < 5$). According to the degree of contamination (C_d) and ecological risk factor (ERF), all stations were categorized as low to moderate contaminated sites ($4.5 < C_d < 17$), and biological communities in some locations may be at risk ($ERF > 65$). Results indicate that application of fertilizers, herbicides and pesticides in agricultural soils has led to soil contamination and special management and educational plans are needed for public and farmers to prevent further adverse effects.

Keywords Agricultural soil · Pollution assessment · Heavy metals · Geochemical indices · Enrichment factor

Introduction

Soil as the main habitat of human and the main source of food production via agriculture should be protected from

adverse effects of different contaminants [1, 2]. During previous decades, soil contamination became a global problem due to rapid industrialization, increasing utilization of pesticides in agriculture and global urbanization [3–5]. Since most of heavy

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metals are bound to soil in natural conditions, it acts as an ultimate sink for discharged heavy metals [6, 7]. Previous studies have proved the hazardous effects of soil pollution to heavy metals [8]. Sources of heavy metals in soils are both geogenic (derived from parent materials) and anthropogenic (derived from human activities) [4, 9, 10]. Anthropogenic sources of soil pollution are related to the manufacturing and agricultural activities like mining, metallurgy, electroplating, fuel combustion, waste disposal, long-term application of sewage sludge, application of fertilizer and irrigation with wastewater [11, 12]. Furthermore, agricultural related activities such as application of chemical fertilizers, pesticides and herbicides followed by the washout of heavy metals and toxic compounds into the soil and water resources can provide a severe threat to the safety of soil and water, since these are two important elements of hydrologic cycle that are directly and indirectly related to each other [13]. Heavy metals accumulation in agricultural soils is a well-known phenomenon and also a major concern, because of their high persistence, toxicity, none-biodegradability, bio-accumulation and biomagnification through the food chain [3, 10]. Entrance of heavy metals into the food chain may pose serious threats to ecosystems and humans [5, 14, 15]. In addition, animals and human exposure to high quantities of heavy metals through soil may pose a serious health risk by ingestion, inhalation and dermal absorption [4]. Long-term application of fertilizers, fungicides and metal-containing pesticides can lead to accumulation of heavy metals in agricultural soils [6, 16, 17]. It should be noted that atmospheric deposition (wet and/or dry) can further increase the levels of soils' heavy metal [18]. The most important parameters for controlling the deposition and bioavailability of heavy metals in soils are physicochemical and biological features of the soil, geochemical characteristics of elements, soil mineralogy, and fertilizer application in agricultural lands and distance to manufacturing areas [18]. Localization of contamination sources and evaluation of agricultural soil according to the distribution of the heavy metals concentration, are considered as effective tools to prevent and control soil contamination [19, 20]. Geostatistical methods are effective tools to discriminate natural sources of contamination from artificial ones [21, 22]. Geostatistical methods can show the spatial distribution and variation of heavy metals in the soil environment. Among various geostatistical methods, Kriging interpolation analysis is a multidimensional method, which can provide valuable data for interpretation of soil pollution level in combination with principal component analysis. It enables examination of the spatial correlation between various variables at multiple scales and determination the source of heavy metals based on the grouping results of the principal component analysis [23–25]. Aghili plain is one of the most critical agricultural centers in Khuzestan province, south west of Iran, in which many industries such as cane factory, metallurgical industries, and paper and pulp

manufacturing are located. Application of chemical fertilizers, pesticides, and herbicides is a possible source of heavy metals emission into the agricultural soil in study area. Since Khuzestan province is a major source of agriculture in Iran, containing several vast agricultural plains and provides a variety of food crops for near 5,000,000 persons, it is very important to evaluate the safety of soil and the agricultural crops in term of toxic elements such as heavy metals. Therefore, investigation of spatial distribution of heavy metals and related health risks is necessary in potentially contaminated soils. Consequently, an intensive observation was conducted through current research for monitoring the concentrations of heavy metals in study area to perform Geo-statistical analysis.

Material and methods

Study area and sample collection

Aghili plain, with 11,000 ha area, is located between 32°1' to 32°7' N and 48°52' to 48°56' E. The climate conditions of this plain are hot and dry (semi-desert) with a rainfall of 326 mm/y and an average temperature of 33 °C. Geologically, the study area is widely characterized by quaternary unconsolidated alluvial sediment (Fig. 1) [26]. The main land use of study area belongs to agricultural lands (80%) followed by national lands with no special use (12%) and the remaining corresponds to residential areas as well as some small industries. The samples were collected by gridding the total area into equal squares, in which one sample was taken per 200 ha. [26]. Afterwards, 54 topsoil samples (0–20 cm) were collected using a stainless steel hand auger in March 2016 (dry season) (Fig. 1) [27]. In each sampling point, a total of 1 ± 0.5 kg of soil was taken from the mixed samples using a quartile method. The collected soil samples were stored in polyethylene bags and then carried to the laboratory [28]. Then, samples were air-dried at room temperature and passed via a 2-mm sieve. Finally, the dried soil samples were sent to the MS analytical Laboratory (Langley, Canada). The soil samples were digested using the Aqua Regia, and the concentration of 12 potentially toxic elements (PTEs) including (As, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, V, Zn, and Hg) were measured using inductively coupled plasma mass spectrometry (ICP-MS, Model: SPECTRO ARCOS, Germany).

Physicochemical characteristics of soil samples

Hydrometer method modified by [29] was applied for finding the size of soil particles and the samples were categorized, based on the amounts of sand, sil and clay. The amount of

Fig. 1 Geological map of Aghili plain and sampling points in the study area

total organic carbon (TOC) of the soil was measured via loss on ignition (LOI) method [30]. In addition, acidity (pH) and salinity of soil samples were determined using mixture of soil and distilled water with 1:2.5 (g:mL) ratio which were shaken during 15 min, prior to determining pH [31, 32].

Quality assurance (QA) and quality control (QC)

Analytical duplicates/replicates, standard reference material (OREAS 24b, and GBM908–10), and blank reagents were used for QA/QC. The average differences of the measured

Table 1 Elemental concentrations in the agricultural soil of Aghili plain

Statistic	As	Cd	Co	Cr	Cu	Mn	Mo	Ni	Pb	V	Zn	Hg	Ref
Minimum (mg/kg)	0.08	0.20	2.60	16.00	7.70	176.00	0.37	23.90	2.40	18.00	13.00	0.01	
Maximum (mg/kg)	6.20	0.54	12.10	60.00	34.40	468.00	2.72	87.10	10.50	45.00	68.00	0.03	
1st Quartile (mg/kg)	1.60	0.27	6.63	35.00	15.80	308.25	0.75	49.23	5.00	31.00	36.00	0.01	
Median (mg/kg)	2.90	0.29	8.10	39.00	17.75	345.00	0.97	58.35	5.90	34.00	42.00	0.01	
3rd Quartile (mg/kg)	3.65	0.33	9.78	44.00	20.28	401.00	1.18	67.50	6.95	37.00	49.00	0.02	
Mean (mg/kg)	2.77	0.30	8.09	39.57	18.39	348.13	1.01	58.66	6.12	33.63	43.09	0.01	
Variance	2.08	0.00	3.76	55.38	20.20	3412.42	0.18	179.18	2.42	20.92	104.24	0.00	
Standard deviation	1.44	0.06	1.94	7.44	4.49	58.42	0.43	13.39	1.56	4.57	10.21	0.01	
Skewness	0.18	1.49	-0.14	-0.27	1.44	-0.28	1.49	0.02	0.86	-0.56	0.21	1.36	
Kurtosis	-0.70	4.52	-0.17	1.17	4.21	0.00	3.79	-0.26	1.19	1.48	0.95	0.87	
CV (%)	52.14	19.66	23.96	18.80	24.44	16.78	42.26	22.82	25.42	13.60	23.69	49.60	
Background	4.7	1.1	6.9	42	14	418	1.8	18	25	60	62	0.1	[41]

*: Coefficients of variation

Note: The sample size: 54

Fig. 2 Spatial distribution of PTEs concentration (mg/kg) in soil samples

and certified values were around 10% variability. The validation parameters of the analytical procedure are presented in Table S1.

Data analysis

All measured data were analyzed using Surfer 14, Excel, XLSTAT, SPSS v.23 software for windows. The original concentration data normality was evaluated using Kolmogorov-Smirnov test (significance level was considered as P value ≤ 0.05). Spearman correlation analysis was used in order to assess the correlation between PTEs. The principal component analysis (PCA) was applied to explicate the relationship and sources of PTEs. In the statistical analysis, those original concentrations lower than the detection limit (DL) were presumed equal to 0.75 of the DL.

Soil quality assessment

For determination of PTEs pollution level in agricultural soil of Aghili plain, some geochemical assessment techniques including enrichment factor (EF), contamination factor (CF), integrated ecological risk index (ERI) and monomial ecological risk factor (ERF) were used. EF is a common approach for determining soil metal sources (anthropogenic or natural events) by normalizing metal concentration, according to soil textural properties and/or un-contaminated background levels [33, 34]. In this study, due to the lack of non-contaminated local background data, the average worldwide soil data (Table 2) was used as reference baselines and Al was considered as a reference element.

EF (unitless) was calculated using following Eq. (1):

$$\text{Enrichment factor} = (C_s/A_s)/(C_B/A_B) \quad (1)$$

Fig. 2 (continued)

Table 2 Concentrations of heavy metals in soil from other studies compared to current work

Study area	Average concentrations of heavy metals (mg/kg)								Ref
	Cr	Ni	Cu	Zn	Cd	Pb	As	Hg	
Yellow River basin, China	79.42	36.05	32.01	117.76	0.46	32.1	11.15	0.14	[35]
Qinghai-Tibet Plateau	93.29	54.73	40.74	145.74	0.68	72.49	—	0.28	[36]
Yerevan, Armenia	66.4	31.4	57.90	195	0.56	2.4	0.69	0.09	[37]
Changsha, China	85.74	—	29.75	139.21	3.10	26.67	—	—	[18]
Krakow, Poland	9.9	6.50	13.40	56.80	0.80	33.9	—	—	[38]
Shushtar, Iran	39.58	58.66	18.39	43.09	0.30	6.12	2.77	0.01	Current work

Table 3 Test of normality for PTEs in agricultural soil of Aghili plain

Element	Kolmogorov-Smimova		
	Statistic	df	Sig.
As	.069	54	0.200*
Cd	.113	54	0.043
Co	.087	54	0.200*
Cr	.085	54	0.200*
Cu	.126	54	0.032
Mn	.083	54	0.200*
Mo	.133	54	0.019
Ni	.059	54	0.200*
Pb	.140	54	0.010
V	.077	54	0.200*
Zn	.094	54	0.200*
Hg	.387	54	0.000

where, C_S , C_B , are the concentrations of element in sample and background (mg/kg), respectively. EF values of 0.5–1.5 and higher than 1.5 reveal the natural and anthropogenic sources, respectively [3]. Furthermore, Enrichment factor values were interpreted as: $EF < 1$ is no enrichment; $1 < EF < 3$ is minor enrichment; $3 < EF < 5$ is moderate enrichment; $5 < EF < 10$ is moderately severe enrichment, $10 < EF < 25$ shows severe enrichment; $25 < EF < 50$ indicate very severe enrichment; and $EF > 50$ is extremely severe enrichment [39].

Table 5 Principal component analysis of PTEs in agricultural soil of Aghili plain

Heavy metal	Component				
	1	2	3	4	5
As	.439	.207	-.491	.639	.021
Cd	.027	.259	.917	-.178	-.003
Co	.911	.335	-.063	.176	-.078
Cr	.822	.490	.223	-.056	-.033
Cu	.328	.911	.038	.066	-.094
Mn	.816	.391	-.115	.261	-.001
Mo	.204	.034	-.181	.950	-.008
Ni	.907	.234	-.016	.140	-.121
Pb	.357	.902	.067	.151	.002
V	.688	.571	.314	.003	.046
Zn	.393	.861	.255	-.087	-.044
Hg	-.080	-.057	-.005	-.004	.993

Extraction Method: Principal Component Analysis.

Rotation Method: Varimax with Kaiser Normalization.

The differences between metal concentrations in soil sample and its own values in background is contamination factor (C_{fi} , unitless). Moreover, the degree of contamination (C_d , unitless) shows the state of metal pollution in soil samples and was calculated using Eq. (2):

$$C_d = \sum C_{fi} = \sum (C_s / C_B) \quad (2)$$

where, C_S , C_B , are the concentrations of element in sample and background (mg/kg). Based on Hakanson's classification, $C_{fi} < 1$ and $C_d < 7$ show low degree of contamination; $1 < C_{fi} < 3$ and $7 < C_d < 17$ imply moderate contamination; $3 < C_{fi} <$

Table 4 Spearman correlation matrix for PTEs in agricultural soil samples of Aghili plain

As	Cd	Co	Cr	Cu	Mn	Mo	Ni	Pb	V	Zn	Hg	
As	1.000	−.422**	.753**	.443**	.481**	.799**	.683**	.683**	.446**	.330*	.191	−.029
Cd		1.000	−.003	.321*	.312*	−.062	−.354**	.022	.390**	.461**	.439**	.020
Co			1.000	.874**	.803**	.898**	.438**	.958**	.746**	.739**	.608**	−.156
Cr				1.000	.891**	.772**	.180	.868**	.815**	.906**	.803**	−.115
Cu					1.000	.766**	.198	.745**	.922**	.882**	.910**	−.142
Mn						1.000	.481**	.800**	.709**	.710**	.538**	−.118
Mo							1.000	.433**	.254	.137	−.012	.031
Ni								1.000	.675**	.660**	.555**	−.196
Pb									1.000	.863**	.906**	−.011
V										1.000	.852**	−.021
Zn											1.000	−.066
Hg												1.000

**correlation is significant at the 0.01 level (2-tailed)

*correlation is significant at the 0.05 level (2-tailed)

Fig. 3 Enrichment factor of PTEs in agricultural soil of study area.

6 and $14 < C_d < 28$ indicate considerable degree of contamination; and $C_f > 6$ and $C_d > 28$ show very high degree of pollution in the sediment [40].

The integrated ecological risk index (ERI, unitless) and monomial ecological risk factor (ERF, unitless) for heavy metals in the sediments were determined using Eq. (3), as follows:

$$ERI = \sum_i ERF^i = \sum_i Tr^i \times Cf^i = \sum_i Tr^i \times (C_s^i / C_n^i) \quad (3)$$

where, Tr^i is the biological toxicity factor for element i , which is defined as $Zn = 1$, $Cr = 2$, $Cd = 30$, $As = 10$, $Hg = 40$ and $Pb = Cu = Ni = 5$ [40]. C_f is contamination factor, and C_s and C_n are metal concentrations in sediments and background values of metal, respectively. The five classes of ERF index are: $ERF < 40$ - low risk; $40 \leq ERF < 80$ - moderate risk; $80 \leq ERF < 160$ - considerable risk; $160 \leq ERF < 320$ - high risk; and $ERF \geq 320$ - very high risk. ERI values categorized as: $ERI < 65$ - low risk; $65 \leq ERI < 130$ - moderate risk; $130 \leq ERI < 260$ - considerable risk; and $ERI \geq 260$ - very high risk [3].

Results and discussions

Grain size and soil characteristics in Aghili plain

According to the findings of hydrometer analysis, the mean values of sand, silt, and clay contents of the studied soils were 18.29%, 42.56%, and 39.15%, respectively. The mean standard deviation values of pH and EC of soil samples of Aghili plain

were 8.12 ± 0.52 and 1.69 ± 0.35 ms/cm, respectively. Moreover, the mean standard deviation of TOC values was 8.24 ± 1.38 . The relatively high organic contents of soil indicate the low bioavailability of PTEs and high formation of organo-metallic complexes [39]. Moreover, the alkaline pH of the studied soil controls the bioavailability (desorption/adsorption and precipitation) of heavy metals. Soil texture in term of grain size can affect the availability of heavy metals. Usually soil with high content of clay and silt tend to sequester heavy metals and vice versa. In this regards, the silt content of 42.56% and clay content of 39.15% demonstrate the high potential studied area in sorption of heavy metals and detrition of soil quality.

Presence and spatial distributions of PTEs

The basic statistical characteristics of the concentrations of 12 PTEs (As, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, V, Zn, and Hg) in agricultural soil samples of Aghili plain are illustrated in Table 1. The mean concentrations of PTEs followed this order: $Mn > Ni > Zn > Cr > V > Cu > Co > Pb > As > Mo > Cd > Hg$. Moreover, the average concentrations of all considered PTEs, except Co, Cu, and Ni, were lower than background (Table 1). According to data observed for skewness, the biggest asymmetry was determined for Cd and Mo followed by Cu, Hg, Pb and V. Based on the coefficient of variation (CV), the anthropogenic import of Cd, Cr, Mn, and V in the study area was low ($CV < 20\%$). While, wide concentration ranges and high CV percentage of the other studied metals, especially As, Mo, and Hg, demonstrated that various anthropogenic sources control soil PTEs contents in the study area. The spatial variation of PTEs in agricultural soils of Aghili plain is

Table 6 Contamination factor (Cf) for heavy metals in agricultural soils of Aghili plain

Sample ID	Cf											
	As	Cd	Co	Cr	Cu	Mn	Mo	Ni	Pb	V	Zn	Hg
1	0.70	0.72	1.17	0.95	1.22	0.84	0.58	3.26	0.40	0.58	0.69	0.29
2	0.43	0.56	0.83	0.69	0.94	0.62	0.46	2.34	0.31	0.47	0.52	0.29
3	0.60	0.66	1.06	0.88	1.14	0.76	0.58	2.99	0.38	0.55	0.65	0.14
4	1.32	0.54	1.49	1.02	1.56	1.12	0.52	3.11	0.63	0.68	0.87	0.14
5	0.62	0.68	1.14	0.93	1.26	0.84	0.61	3.14	0.39	0.57	0.71	0.11
6	0.36	0.56	0.84	0.74	1.01	0.69	0.51	2.42	0.33	0.48	0.58	0.14
7	0.62	0.72	1.45	1.12	1.47	1.02	0.53	3.78	0.50	0.67	0.84	0.14
8	0.02	0.52	0.38	0.38	0.55	0.42	0.39	1.33	0.16	0.30	0.21	0.11
9	0.43	0.68	1.13	1.05	1.44	0.88	0.55	3.21	0.39	0.63	0.76	0.11
10	0.64	0.64	1.17	1.05	2.46	0.88	0.70	3.26	0.70	0.60	1.08	0.14
11	0.30	0.58	0.90	0.74	1.05	0.72	0.57	2.47	0.32	0.50	0.53	0.14
12	0.74	0.60	1.19	0.90	1.28	0.74	0.58	3.47	0.50	0.57	0.71	0.29
13	0.45	0.94	1.59	1.43	1.90	0.99	0.22	4.19	0.59	0.75	1.10	0.11
14	0.28	0.70	0.94	0.81	1.13	0.72	0.54	2.71	0.33	0.53	0.66	0.14
15	0.30	0.62	1.03	0.93	1.24	0.77	0.33	2.90	0.39	0.57	0.68	0.14
16	0.64	0.64	1.17	1.02	2.44	0.88	0.65	3.27	0.70	0.60	1.08	0.14
17	0.34	0.60	0.93	0.83	1.13	0.70	0.47	2.72	0.35	0.53	0.60	0.29
18	0.15	0.58	0.87	0.74	1.09	0.64	0.41	2.51	0.32	0.47	0.61	0.14
19	0.17	0.56	0.81	0.74	1.08	0.63	0.41	2.37	0.32	0.47	0.65	0.14
20	0.49	0.58	0.81	0.79	1.08	0.78	0.42	2.04	0.33	0.52	0.50	0.43
21	0.55	0.70	1.22	1.02	1.41	0.84	0.88	3.41	0.47	0.63	0.79	0.29
22	0.34	0.68	1.04	0.88	1.19	0.74	0.41	3.02	0.39	0.55	0.68	0.11
23	0.43	0.60	0.91	0.86	1.27	0.70	0.49	2.65	0.40	0.53	0.82	0.14
24	0.15	0.70	1.19	1.14	1.40	0.75	0.21	3.55	0.43	0.63	0.89	0.29
25	0.57	0.54	0.96	0.81	1.14	0.88	0.43	2.34	0.35	0.57	0.58	0.14
26	0.13	0.56	0.68	0.57	0.80	0.57	0.43	2.07	0.25	0.40	0.40	0.43
27	0.21	0.68	1.13	1.07	1.39	0.76	0.24	3.23	0.40	0.62	0.74	0.14
28	0.26	0.70	1.19	1.07	1.33	0.76	0.24	3.47	0.42	0.62	0.81	0.14
29	0.51	0.42	1.01	0.90	1.14	0.78	0.62	2.77	0.35	0.57	0.60	0.14
30	0.19	0.64	0.93	0.86	1.15	0.71	0.32	2.77	0.34	0.53	0.68	0.14
31	0.26	1.08	0.99	0.93	1.29	0.78	0.23	2.82	0.41	0.55	0.74	0.11
32	0.64	0.72	1.36	1.12	1.57	0.96	0.44	3.49	0.51	0.67	0.92	0.43
33	0.51	0.50	0.97	0.81	1.04	0.74	0.62	2.53	0.32	0.50	0.55	0.29
34	0.13	0.52	0.84	0.79	1.09	0.72	0.34	2.42	0.32	0.50	0.56	0.14
35	0.68	0.64	1.26	1.02	1.46	0.96	0.53	3.09	0.47	0.63	0.82	0.29
36	1.06	0.42	1.09	0.83	1.26	0.83	0.32	3.05	0.31	0.48	0.56	0.14
37	0.74	0.58	1.49	1.12	1.32	0.98	0.79	4.23	0.40	0.62	0.66	0.11
38	0.64	0.64	1.62	1.24	1.60	0.89	0.40	4.84	0.45	0.63	0.84	0.11
39	1.02	0.50	1.51	1.12	1.55	1.00	0.55	4.29	0.47	0.58	0.79	0.29
40	1.13	0.54	1.65	1.19	1.41	1.06	0.66	4.68	0.41	0.63	0.71	0.14
41	0.66	0.50	1.12	0.76	1.11	0.79	0.49	3.06	0.33	0.45	0.55	0.11
42	1.09	0.40	1.49	1.00	1.50	1.00	0.87	3.76	0.52	0.58	0.76	0.14
43	0.81	0.42	1.20	0.90	1.13	0.82	0.68	3.43	0.32	0.52	0.53	0.29
44	0.72	0.62	1.57	1.12	1.60	0.95	0.46	4.23	0.49	0.60	0.87	0.11
45	1.04	0.54	1.75	1.21	1.71	1.03	0.66	4.68	0.59	0.65	0.94	0.14
46	0.74	0.58	1.20	0.95	1.11	0.94	0.55	3.53	0.37	0.52	0.58	0.29
47	1.00	0.60	1.41	0.98	1.45	0.98	1.51	3.69	0.51	0.60	0.71	0.14

Table 6 (continued)

Sample ID	Cf											
	As	Cd	Co	Cr	Cu	Mn	Mo	Ni	Pb	V	Zn	Hg
48	1.02	0.54	1.46	1.05	1.47	0.97	1.13	3.97	0.45	0.62	0.71	0.14
49	0.79	0.48	1.29	0.93	1.18	0.82	0.68	3.73	0.33	0.52	0.55	0.11
50	0.89	0.50	1.51	1.02	1.42	1.04	0.74	4.18	0.42	0.55	0.73	0.14
51	0.83	0.56	1.33	0.93	1.23	0.91	0.67	3.81	0.37	0.53	0.58	0.11
52	0.89	0.44	1.16	0.83	1.11	0.80	0.87	3.31	0.35	0.48	0.55	0.11
53	0.83	0.56	1.42	1.02	1.29	0.96	1.06	4.11	0.41	0.58	0.66	0.29
54	0.74	0.52	1.48	1.07	1.32	0.94	0.65	4.27	0.37	0.58	0.66	0.11

illustrated in Fig. 2. The obtained results demonstrated that the west and northwest parts of the study area have lower contents of PTEs. Similar spatial distribution trends for Co, Cr, Mn, V and Ni; Pb, Zn, and Cu indicated the same sources for mentioned PTEs in each group. Cr, Co, Mn, V, and Ni are mainly

from natural sources. Animal manure, commercial fertilizers, fungicides and pesticides contain high amounts of Zn, and Cu [4, 39]. Moreover, fuel combustion of agricultural machinery could also be considered as another source of Pb, Zn, and Cu in the study area. According to Fig. 2, the spatial variation of

Fig. 4 **a** Variations of degree of contamination (C_d) of heavy metals in different samples of agricultural soils **(b)** spatial distribution C_d in study area and

Table 7 The integrated ecological risk index (ERI) for heavy metals in agricultural soils of Aghili plain

Stations	ERI							
	Zn	Cr	Cd	As	Hg	Pb	Cu	Ni
1	0.69	1.90	21.60	7.02	11.43	2.00	6.11	16.28
2	0.52	1.38	16.80	4.26	11.43	1.57	4.71	11.72
3	0.65	1.76	19.80	5.96	5.71	1.90	5.71	14.97
4	0.87	2.05	16.20	13.19	5.71	3.17	7.82	15.56
5	0.71	1.86	20.40	6.17	4.29	1.97	6.32	15.69
6	0.58	1.48	16.80	3.62	5.71	1.67	5.04	12.08
7	0.84	2.24	21.60	6.17	5.71	2.50	7.36	18.92
8	0.21	0.76	15.60	0.16	4.29	0.80	2.75	6.64
9	0.76	2.10	20.40	4.26	4.29	1.97	7.21	16.06
10	1.08	2.10	19.20	6.38	5.71	3.50	12.29	16.31
11	0.53	1.48	17.40	2.98	5.71	1.60	5.25	12.36
12	0.71	1.81	18.00	7.45	11.43	2.50	6.39	17.36
13	1.10	2.86	28.20	4.47	4.29	2.97	9.50	20.94
14	0.66	1.62	21.00	2.77	5.71	1.63	5.64	13.56
15	0.68	1.86	18.60	2.98	5.71	1.97	6.21	14.50
16	1.08	2.05	19.20	6.38	5.71	3.50	12.21	16.33
17	0.60	1.67	18.00	3.40	11.43	1.73	5.64	13.61
18	0.61	1.48	17.40	1.49	5.71	1.60	5.43	12.53
19	0.65	1.48	16.80	1.70	5.71	1.60	5.39	11.83
20	0.50	1.57	17.40	4.89	17.14	1.63	5.39	10.22
21	0.79	2.05	21.00	5.53	11.43	2.37	7.07	17.06
22	0.68	1.76	20.40	3.40	4.29	1.97	5.96	15.11
23	0.82	1.71	18.00	4.26	5.71	2.00	6.36	13.25
24	0.89	2.29	21.00	1.49	11.43	2.17	7.00	17.75
25	0.58	1.62	16.20	5.74	5.71	1.73	5.68	11.72
26	0.40	1.14	16.80	1.28	17.14	1.23	4.00	10.33
27	0.74	2.14	20.40	2.13	5.71	2.00	6.93	16.14
28	0.81	2.14	21.00	2.55	5.71	2.10	6.64	17.33
29	0.60	1.81	12.60	5.11	5.71	1.73	5.68	13.86
30	0.68	1.71	19.20	1.91	5.71	1.70	5.75	13.86
31	0.74	1.86	32.40	2.55	4.29	2.03	6.46	14.11
32	0.92	2.24	21.60	6.38	17.14	2.53	7.86	17.47
33	0.55	1.62	15.00	5.11	11.43	1.60	5.18	12.64
34	0.56	1.57	15.60	1.28	5.71	1.60	5.46	12.08
35	0.82	2.05	19.20	6.81	11.43	2.37	7.29	15.47
36	0.56	1.67	12.60	10.64	5.71	1.53	6.32	15.25
37	0.66	2.24	17.40	7.45	4.29	2.00	6.61	21.14
38	0.84	2.48	19.20	6.38	4.29	2.27	8.00	24.19
39	0.79	2.24	15.00	10.21	11.43	2.33	7.75	21.44
40	0.71	2.38	16.20	11.28	5.71	2.07	7.04	23.42
41	0.55	1.52	15.00	6.60	4.29	1.67	5.57	15.31
42	0.76	2.00	12.00	10.85	5.71	2.60	7.50	18.78
43	0.53	1.81	12.60	8.09	11.43	1.60	5.64	17.17
44	0.87	2.24	18.60	7.23	4.29	2.43	8.00	21.17
45	0.94	2.43	16.20	10.43	5.71	2.97	8.57	23.39
46	0.58	1.90	17.40	7.45	11.43	1.87	5.54	17.64

Table 7 (continued)

Stations	ERI							
	Zn	Cr	Cd	As	Hg	Pb	Cu	Ni
47	0.71	1.95	18.00	10.00	5.71	2.53	7.25	18.47
48	0.71	2.10	16.20	10.21	5.71	2.23	7.36	19.83
49	0.55	1.86	14.40	7.87	4.29	1.67	5.89	18.67
50	0.73	2.05	15.00	8.94	5.71	2.10	7.11	20.89
51	0.58	1.86	16.80	8.30	4.29	1.83	6.14	19.06
52	0.55	1.67	13.20	8.94	4.29	1.73	5.57	16.53
53	0.66	2.05	16.80	8.30	11.43	2.07	6.46	20.56
54	0.66	2.14	15.60	7.45	4.29	1.83	6.61	21.36

Hg, Cd, As and Mo completely differed from the others. Since the hotspot of Hg is close to the residential area, human activities could be the sources of Hg in the study area. Moreover, Cd, usually presents in all agricultural soils, due to wide application of manure and phosphorous fertilizers [42]. Table 2 presents the concentrations of heavy metals in soil obtained for other sites in the world like China, Poland and Armenia compared to current work. As can be seen, the concentrations vary widely depending on the kind of heavy metal and also study area. But, some similarities can be seen in the trends of variations between the kinds of heavy metals. For example the most values belong to Zn and the least to the Hg, almost for all samples from different studies.

Source identification of PTEs in soil

The results of Kolmogorov-Smirnov test showed that Cd, Cu, Mo, Pb, and Hg were not normal (Table. 3). Hence, for finding the correlation between PTEs and their probable origin, the non-parametric spearman correlation analysis was done (Table. 4). A strong positive relationship between Zn, Pb, and Cu ($P < 0.01$, $r > 0.9$) indicated the same sources of contamination (mainly anthropogenic). Moreover, the significant positive relationship was discovered between Co, Cr, Mn, Ni, V ($P < 0.01$, $r > 0.7$), Mo and As ($P < 0.01$, $r > 0.6$) that indicated the similar sources. Cd shows a relatively moderate positive correlation with V and Zn ($P < 0.01$, $r < 0.5$) and weak positive correlation with Cu, Pb, Cr ($P < 0.01$, $r < 0.4$). Furthermore, the relationship between Cd, As and Mo, was negative ($P < 0.01$, $r = -0.422$ and $r = 0.354$, respectively). Finally, an insignificant relationship was observed between Hg and the other PTEs ($P > 0.05$).

In order to find more information about the relationship between heavy metals and their sources, principal component analysis (PCA) was carried out (Table 5). The results of applied PCA were completely in line with correlation analysis. The five principal components (PCs) showed 94.65% of the variance within the data set. The first factor displayed 34.97%

of the total variance including Co, Cr, Mn, Ni, and V. The grouping of Ni and V mostly indicates the crude oil sources as well as transportation [33, 43]. This group of elements suggests mixed sources of contamination (both anthropogenic and natural). The second PC accounting for 28.21% of the total variance indicates loading of Pb, Cu, and Zn, showing the role of anthropogenic sources (combustion of fossil fuels and fertilizers). Proximity of two highway and several local ways contribute in emission of heavy metals such as lead due to combustion of fossil fuels [39, 44]. The third group shows 12.2% of the total variance and dominated by As and Mo (mainly from natural sources). Factor 4 displays 9.69% of the total variance and defined by Cd. Phosphorous fertilizers, pesticides and municipal solid wastes are the main sources of Cd in agricultural soil [4, 16, 39, 42]. Finally, the fifth PC was accounted for 8.50% of the total variance includes Hg (both human activities and natural sources). Hence, the natural and chemical fertilizers and human activities are the most important sources of soil pollution in the study area. It should be noted that the majority of land use in study area was agricultural soil and application of herbicides and pesticides as well as chemical fertilizers have the highest contribution in land pollution to heavy metals. Of course, other pollution sources such as industrial plants and transportation should not be neglected [44].

Quality of the agricultural soil

Figure 3 demonstrates the calculated EF values of PTEs in agricultural soil of Aghili plain. The EF values of all studied elements in either all or some stations were higher than 1.5, indicating the influence of human activities in the study area. However, EF values of all metals, except Ni, were in the range of non to moderate enrichment. While, EF values for Ni, in most stations, indicated severe enrichment ($10 < EF < 25$). The probable reason for soil enrichment with oil-associated elements

(Ni, Mn, V, Zn, and Co) in the study area is the application of kerosene as herbicides.

The calculated values of Cf are presented in Table 6. Results indicated that Cf values of As, Cd, Cr, Mn, Mo, Pb, V, Zn and Hg, in most stations, were within the range of low degree of contamination ($Cf < 1$). While, Cf values of Co and Cu were in the range of moderate contamination ($1 < Cf < 3$) and Cf value of Ni was in the range of considerable contamination ($3 < Cf < 6$). Moreover, the variations of C_d in different sample points is presented in Fig. 4a. As is shown in Fig. 4b and based on calculated values of C_d , all stations were categorized as low to moderate contaminated sites ($4.5 < C_d < 17$).

Based on the calculated values of the ERI (Table 7), the agricultural soils of Aghili plain were classified as low potential ecological risk ($ERI < 40$). However, considering the ERF values (Fig. 5), the biological communities in some stations may be at risk ($ERF > 65$). Results are in accordance with study by Jorfi et al., 2017 on heavy metals pollution in Mianab plain, Dezful city, Iran [34].

Conclusion

Spatial distribution of some important heavy metals were investigated in agricultural soils of Aghili plain, in south weste of Iran. In this regards, soil quality assessment was performed using geochemical indices. The EF values of higher than 1.5 proved the influence of human activities. Principal component analysis (PCA) indicated that the chemical fertilizers, transportation, drainages containing herbicides and pesticides are the most important sources of heavy metals emission to soil. ERI values indicated that the agricultural soils of Aghili plain were classified as low potential ecological risk, but, the biological communities in some stations may be at risk ($ERF > 65$). This demonstrated the serious requirement for planning and conducting management strategies in order to maintain the soil quality and mitigate the adverse effects of human

Fig. 5 Monomial ecological risk factor (ERF) for heavy metals in agricultural soils of Aghili plain

activities in terms of heavy metals entrance to agricultural soils as well as subsequent human health risks associated with exposure to heavy metal through contaminated soils and agricultural crops. Training plans for farmers, replacing chemical fertilizers and pesticides with harmless ones, implementation of perfect environmental impact assessment for industries and periodical sampling programs would be effective in keeping and promoting soil quality.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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