Journal of Environmental Health Science and Engineering (2021) 19:107–120
https://doi.org/10.1007/s40201-020-00585-7

RESEARCH ARTICLE

Bioavailability, distribution and health risk assessment of arsenic and heavy metals (HMs) in agricultural soils of Kermanshah Province, west of Iran

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Received: 13 December 2019 / Accepted: 10 November 2020 / Published online: 6 January 2021

Abstract

Kermanshah Province as an agricultural hub exports food crops to neighboring countries. In this study, contamination status, bioavailability, spatial distribution, and ecological and human health risk of arsenic and heavy metals (HMs) in soil were investigate. For this purpose, 121 agricultural soil samples were collected and analyzed using ICP-MS. The data were studied by calculating some geochemical indices, and using geographical information system and statistical analysis. Results showed that Cd has the highest bioavailability, following by Cu and As. Also, Cu was severely associated with organic matter. Enrichment factor (EF) followed the order of As > Cu > Pb > Se > Cd > Zn > Ni > Cr, and the soil pollution index (SPI) ranged from 0.82 to 2.65. Low potential ecological risk was measured for most of the samples. However, Kermanshah County and Eastern parts of the Province showed the highest HMs enrichment and ecological risk. Moreover, high carcinogenic risk of Cr and Ni threatens the children. Cr showed also high non-carcinogenic hazard index (HI) for children. Principal component analysis (PCA) indicated the anthropogenic origins for As, Cd, Cu, Pb, Se and Zn, while Cr and Ni originated mainly from a geogenic source. Furthermore, Kruskal-Wallis H test revealed that As, Cd, Cr, Cu, Ni, Pb, Se and Zn concentrations were significantly different ($p < 0.05$) between 16 Counties of the Kermanshah Province. Overall, the management of urban and industrial contamination sources is required to minimize the concentration of bioavailable portion of HMs and preventing residents of the area from being exposed to contaminants.

Keywords Bioavailability · Sequential extraction · Heavy metals · Soil · Health risk · Kermanshah

Introduction

Human activities including mineral exploitation, food and industrial processing, and agricultural, domestic and commercial activities have an important role in making the environment harmful to the humans and other organisms. As one of the direct consequences, arsenic and heavy metals (HMs) will be deposited on top soils and water bodies [1, 2]. Soil is the basis of terrestrial ecosystems and the medium for occurring various biogeochemical cycles. Trace elements in the environment are persistent and could be toxic and bioaccumulated, so soil contamination by toxic elements is dangerous and has attracted more attention [3].

Numerous studies in the world focused on agricultural soils HMs, including spatial distribution, pollution level, identification of potential sources, food security and accumulation characteristics [4–8] have been reported in recent years. In Iran as well, the assessment of heavy metal pollution in agricultural soil and its effect on the environment and ecology has been conducted in several studies [9–13]. Agricultural soil have two HMs sources including natural (weathering, erosion of parent rocks, atmospheric deposition and volcanic activities, etc.) and anthropogenic (sewage irrigation, addition of

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manures, fertilizers and pesticides, etc.) sources, both are responsible for soil and crops contamination [14–16].

Some heavy metals such as Cu, Zn and Fe have been identified as essential elements for crop growth. However these micronutrients could be phytotoxic in higher concentrations. Some other such as Co and Se are not required by plant growth but are essential for animals. Also, some HMs including Ni, As, Cd, Pb, Cr and Hg have not been identified as essential elements for plants or animals and even at low concentrations may be severely toxic to living organisms [17]. Accumulation of HMs in agricultural soil may cause loss of soil nutrients and soil function degeneration, which consequently decreases the crops quality. Moreover, they will cause negative health effects on human in the long term through food chain due to their high toxicity, long residence times and accumulation in agricultural soils [14, 17–21]. However, HMs accumulate in different forms in soil, and their total concentration could not represent the bioavailable fraction for food crops. Therefore, as the most popular methods, sequential extraction procedures and extraction with a solution of a complexing agent, such as ethylene-diamino-tetra acetic acid (EDTA), are used to evaluate mobility and bioavailability of elements in soil [22, 23]. On the other hand, agricultural soils are loose due to continuous plowing and can travel long distances as a result of wind. Therefore, in addition to the farmers, the inhabitants of the remote areas are also exposed to these soils, via inhalation, ingestion, and dermal routes.

Kermanshah Province is a representative area showing rapid population. Owing to the vast crop lands and unique natural environment, Kermanshah is one of the agricultural centers in western part of Iran, and agriculture acts the major role in its economy [24]. The excess application of fertilizers in this area may cause an increase in HMs contents of agricultural soil [25], which in turn can result ecological and health risks for inhabitants and consumers of grown food crops in these lands. Based on the above background, the main objectives of this study were to (a) assess the contamination level and bioavailability of HMs; (b) identify the main pollution sources; (c) evaluate the spatial variability; and (d) assess the ecological risk and the health risk of HMs via inhalation, ingestion, and dermal routes in agricultural soils of Kermanshah Province.

Materials and methods

Study area

Kermanshah province, as an important agricultural area in Iran, is located in West of Iran, between 33° and 35° latitudinal and 45° and 48° longitudinal geographical coordinates (Fig. 1). It has a total area of approximately 25,008 km^2 with a population of over 1,945,227 inhabitants [26]. Mean annual precipitation in the study area is about 403.6 mm, and annual mean temperature is 15.76 °C (for the period of 2010 to 2017) [27]. Based on Geological Maps, the study area is mostly covered with limestone, dolomite, sandstone, shale and evaporates, and limited outcrops of gabbro, basalt, peridotite, andesite and granite (Fig. 1). The soils in the study area are mostly composed of inceptisols, vertisols, entisols and badlands [28].

Sample collection, preparation and analysis

At the harvest time, a total of 121 agricultural (at 0–15 cm in depth), and 11 pristine soil samples (for geochemical baseline, at 30–45 cm in depth) were collected using a plastic scoop. To achieve a representative sample, composite samples were prepared by mixing five subsamples. The location of collected soil samples have been showed in Fig. 1. In the laboratory, the samples were air-dried at ambient temperature and for the determination of heavy metals sieved through a 63-μm sieve. Also, the samples were sieved to 2 mm for the rest of the physicochemical parameters. The total concentrations of nine metal/loids (Al, As, Cd, Cr, Cu, Ni, Pb, Se and Zn) were measured in an accredited commercial laboratory (Zar Azma Laboratory, Iran) using ICP-MS methods (Agilent, 7700x, USA). Before analyses, approximately 1 g of each sample was extracted using HF, HNO_3, $HClO_4$ and H_2O_2 mixture in a Teflon beaker. Internal duplicates, blanks, and HRM were used to data quality assurance and control. The measurements accuracy and precision are ± 4% and 96%, respectively. For bioavailability evaluations, soil samples from four zones (North and center, East, West, and South of the Province) were chosen. Estimation of plant-available metals was carried out by extraction with 0.05 M EDTA at pH 7 [29], and BCR protocol modified by Mossop and Davidson [30] was used for sequential extraction. The residual fraction was extracted by the same method as in total metal digestion, and the average recovery percentages ranged from 81.6 to 113.5 for studied elements.

Soil pH, electrical conductivity (EC) and cation exchange capacity (CEC) were determined according to Ryan et al. [31]. The samples were analyzed to determine organic matter (OM) by the LOI procedure [32], and particle size distribution (sand, silt, and clay content) was determined using the hydrometer method. Geochemical baselines for studied metals were measured using median absolute deviation method [33, 34].

Data analysis

Enrichment factor (EF)

Enrichment factor (EF) is widely used as an appropriate approach to discriminate between natural and anthropogenic sources and to reflect the status of environmental contamination, based on the use of a normalization element in order to

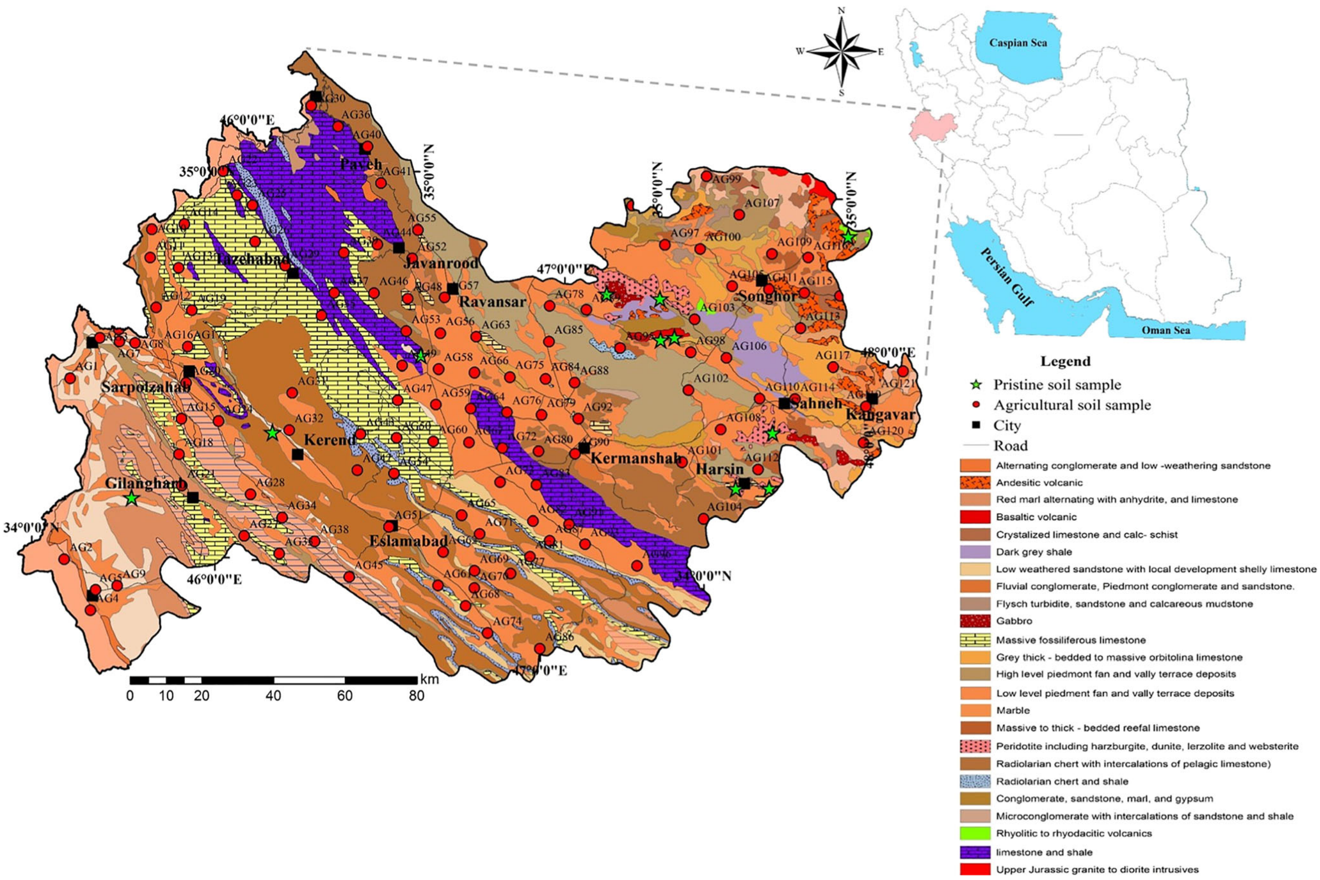

Fig. 1 Geological map, and location of sampling stations

alleviate the variations produced by heterogeneous sediments [35]. The calculation equation is listed as follows [36]:

$$\mathrm{EF} = (\mathrm{X}/\mathrm{Al})_{\mathrm{sample}}/(\mathrm{X}/\mathrm{Al})_{\mathrm{Background}} \tag{1}$$

Where X refers to the concentration of certain heavy metal in the samples or in Earth crust. In this study, Al was used as normalizing element to calculate the enrichment factor. EF < 1 indicates no enrichment, 1–3 minor enrichment, 3–5 moderate enrichment, 5–10 moderate to severe enrichment, 10–25 severe enrichment, 25–50 very severe enrichment, and > 50 extremely severe enrichment [37].

Soil pollution index (SPI)

To better demonstrate the spatial distribution of enriched HMs, soil pollution index was calculated at each sampling station as follows [38, 39]:

$$\mathrm{SPI}_{\mathrm{i}} = \frac{\sum_j \frac{MCi}{TCj}}{N} \tag{2}$$

Where i = the sampling stations, j = the enriched HMs, MC_{i} = the metal concentrations at the sampling station, TC_{j} = the geochemical baseline of j_{th} metals that are enriched, and N is the number of enriched trace metals.

Potential ecological risk index (PER)

The potential ecological risk index (PER) which was also presented to assess the contamination degree of heavy metals in sediments/soils, could be calculated as follows [40]:

$$\mathrm{PER} = \sum \mathrm{E} \tag{3}$$

$$\mathrm{E} = \mathrm{TC} \tag{4}$$

$$\mathrm{C} = \mathrm{C}_{\mathrm{a}}/\mathrm{C}_{\mathrm{b}} \tag{5}$$

Where C is the single HM pollution factor, C_{a} is the content of the metals in samples, and C_{b} is the geochemical baseline of the metal. PER is a comprehensive potential ecological index, E is the ecological risk of individual metals or potential risk factor, T is toxic response factor which for the analyzed elements is taken as Zn = 1 < Cr = 2 < Cu = Ni = Pb = 5 < As = 10 < Cd = 30. The potential risk factors are classified as: low

($E < 40$); moderate ($40 \leq E < 80$); considerable ($80 \leq E < 160$); high ($160 \leq E < 320$); and very high ($E \geq 320$). Consequently the potential ecological risk is: low (PER <150); moderate ($150 \leq$ PER <300); considerable ($300 \leq$ PER <600); or very high (PER ≥600) [34, 41].

Health risk assessment

In order to evaluate the health risks (carcinogenic and non-carcinogenic) posed by HMs in the studied soils, the risk assessment model developed by the Environmental Protection Agency of the United States was used. Average daily doses (ADD) received through inhalation, dermal contact and ingestion could be calculated as follows [34, 42, 43]:

$$\mathrm{ADD_{ing}} = \frac{HM \times IR_{ing} \times \mathrm{EF} \times \mathrm{ED} \times 10^{-6}}{BW \times \mathrm{AT}} \tag{6}$$

$$\mathrm{ADD_{inh}} = \frac{HM \times IR_{inh} \times \mathrm{EF} \times \mathrm{ED}}{PEF \times BW \times \mathrm{AT}} \tag{7}$$

$$\mathrm{ADD_{dermal}} = \frac{HM \times SA \times \mathrm{AF} \times \mathrm{ABF} \times \mathrm{EF} \times \mathrm{ED} \times 10^{-6}}{BW \times \mathrm{AT}} \tag{8}$$

ADD means a dose rate averaged over a pathway-specific period of exposure expressed as a daily dose on a per-unit-body-weight basis. The values and definitions of exposure factors are presented in Table 1. The hazard quotient (HQ), hazard index (HI) and cancer risk are subsequently calculated by dividing/multiplying the ADD values for each exposure pathway by corresponding reference dose (RfD)/slope factor (SF), respectively [44]:

$$\mathrm{HI} = \sum \mathrm{HQ_i} = \sum \frac{ADD_i}{RfD_i} \tag{9}$$

$$\mathrm{CR} = \sum \mathrm{ADD_i} \times \mathrm{SF_i} \tag{10}$$

A Hazard Index (HI) of <1, and > 1indicate no adverse and possible adverse health effects, respectively [41]. Also, $CR < 1 \times 10^{-6}$ shows negligible carcinogenic risk, while $1 \times 10^{-6} < CR < 1 \times 10^{-4}$ and $CR > 1 \times 10^{-4}$ reveal tolerable risk and high risk of developing cancer, respectively [44–46].

Statistical and geostatistical analysis

Statistical analysis of the data was carried out using SPSS 19.0 for windows. In this study, multivariate statistical technique, principal component analysis (PCA), was performed for the data set to reveal the relationship between parameters and better source identification. Also, the Kruskal-Wallis H test which is often considered the nonparametric test equivalent to the one-way ANOVA, and an extension of the Mann-Whitney U test to allow the comparison of more than two independent groups, was used to compare HMs concentrations between soil samples of different counties of Kermanshah Province.

Estimation and mapping of soil attributes in un-sampled areas is main application of geostatistics in soil science [47–49]. Geostatistical Analyst tool for ArcMap (ArcGIS 10) was used to choose the best fit method among several spatial interpolation techniques, including radial basic functions (RBF), inverse distance weighting (IDW), local polynomial interpolation (LPI) etc., to display the spatial distribution

Table 1 Exposure factors for risk assessment models

Factor	Definition	Unit	Value	
			Children	Adults
HM	Heavy metal concentration (95% UCL in this study)	mg/kg		
IR_{ing}	Ingestion rate	$mg\ d^{-1}$	100 [61]	50 [61]
IR_{inh}	Inhalation rate	$m^3\ d^{-1}$	7.6 [62, 63]	20 [62, 63]
PEF	Particle emission factor	$m^3\ kg^{-1}$	1.36×10^9 [45]	1.36×10^9 [45]
SA	Exposed skin area	cm^2	2699 [61]	3950 [61]
AF	Skin adherence factor	$mg\ cm^{-2}\ d^{-1}$	0.2 [45]	0.07 [45]
ABF	Dermal absorption factor	–	0.03 [61]	0.001 [61]
ED	Exposure duration	year	6 [45]	24 [45]
EF	Exposure frequency	$d\ y^{-1}$	350 [44]	350 [44]
BW	Average body weight	kg	18.6 [61]	80 [61]
AT	Average life span for heavy metals	d	ED × 365 [64]	ED × 365 [64]

of HMs, SPI and potential ecological risk in agricultural soils of the study area.

Results and discussion

Physicochemical parameters and total metals concentration

Table 2 provides the descriptive statistics of HMs concentration and physicochemical parameters in agricultural soil samples as well as their calculated geochemical baseline in the study area. Results showed that based on USDA ternary diagram, soil texture could be classified as sandy loam, loam and clayey loam (in most of the samples), silty loam, and silty clayey loam, showing the dominance of fine texture. The soil pH ranged from 7.15 to 7.96 with a mean value of 7.69, indicating neutral to slightly alkaline nature of the soils [50]. The soil pH range is ideal for most crops and in some cases free carbonate could be present in soil [31]. Soils CEC values varied from 17 to 88.62% (mean 28.39%) revealing medium to very high value for agricultural soils [51]. Electrical conductivity of the samples ranged from 0.30 to 10.03 mS/cm with an average of 1.37 and 13.36 mS/cm, revealing non-saline to severely saline properties for agricultural soils [52]. Also, mean organic matter content of soils was 2.21% (ranging from 0.06 to 55%), showing a wide range from very low to very high organic matter contents based on Metson [51].

From the Table 2, Al (with a mean concentration of 4.56%) was the most abundant metal in all the samples, because this metal is common element in the Earth crust [53]. The ranges of the concentrations of HMs were as follows: As 2.76–25.51 mg/kg, Cd 0.12–2.50 mg/kg, Cr 40–485 mg/kg, Cu 8.20–46.50 mg/kg, Ni 50.60–474.90 mg/kg, Pb 6.70–18.90 mg/kg, Se 0.07–1.59 mg/kg and Zn 39.40–127 mg/kg. The relatively same concentration ranges were reported for Cu (10–83 mg/kg), Ni (48–306 mg/kg), Cr (32–235 mg/kg) and Zn (40–113 mg/kg) by Doabi et al. [10] in agricultural soils of Kermanshah previously.

The mean concentrations of aluminum, nickel and chromium were lower compared with calculated geochemical baseline values of HMs in the study area, while the rest of the studied HMs showed higher average concentrations. However, all the studied elements had higher concentrations in agricultural soils than the baseline values at least at some sampling stations. Comparison of the results with soil quality guidelines for the protection of environmental and human health [54–57] revealed that the concentrations of As, Cd, Cr, Ni and Se exceeded the guideline values in 2.50% (three samples), 1.65% (one sample), 95.86% (116 samples), 100% (121samples) and 20.66% (25 samples), respectively. It should be noted that Ni and Cr concentrations in geochemical baseline were higher than the CCME guidelines, indicating high background contents and probably a geogenic source of these HMs in the soils of the study area. The highest concentrations of Cr and Ni in agricultural soils were measured at areas with basalt, gabbro and shale outcrops confirming their

Table 2 Descriptive statistics of total HMs concentrations and physicochemical parameters

N = 121	Mean	Median	Std. Deviation	Skewness	Min	Max	Geochemical baseline	Guideline for agricultural soils
Al (%)	4.56	4.54	0.92	−0.05	2.18	7.15	5.09	–
As (mg/kg)	6.93	6.32	2.66	3.16	2.76	25.51	5.20	12 [54]
Cd (mg/kg)	0.39	0.34	0.26	5.43	0.12	2.50	0.42	1.40 [55]
Cr (mg/kg)	115.76	106.00	54.90	3.33	40.00	485.00	148	64.00 [54]
Cu (mg/kg)	20.82	18.25	7.87	1.11	8.20	46.50	19.20	63.00 [55]
Ni (mg/kg)	148.06	125.95	85.32	1.90	50.60	474.90	150	45.00 [57]
Pb (mg/kg)	11.45	11.15	2.50	0.55	6.70	18.90	9.85	70.00 [55]
Se (mg/kg)	0.69	0.60	0.33	0.68	0.07	1.59	0.63	1.00 [56]
Zn (mg/kg)	72.59	72.35	14.72	0.52	39.40	127.00	66.50	200 [55]
Sand (%)	30.98	30.00	9.51	0.67	14.00	60.00	–	–
Silt (%)	44.74	46.00	8.18	−0.49	18.00	64.00	–	–
Clay (%)	23.94	23.00	6.70	0.27	8.00	40.00	–	–
CEC (meq/100 g)	28.39	27.36	9.01	3.44	17.00	88.62	–	–
pH	7.69	7.70	0.13	−0.43	7.15	7.96	–	–
OM (%)	2.21	0.86	6.34	6.20	0.06	55.00	–	–
EC (mS/cm)	1.37	1.13	1.23	4.74	0.30	10.03	–	–

Table 3 EDTA-extractable and percentages of metals content in each steps of sequential extraction

	As	Cd	Cr	Cu	Ni	Pb	Se	Zn
Zone 1 (North and center)								
EDTA extractable (mg/kg)	2.8	0.74	6.72	12.72	9.36	5.89	0.06	28.48
F1%	11.37	49.36	9.71	2.11	2.47	3.53	2.04	4.89
F2%	28.05	22.45	13.28	1.31	1.95	19.84	13.83	28.66
F3%	23.7	15.39	17.51	58.39	8.96	29.51	15.27	26.31
F4%	36.88	12.8	59.5	38.19	86.62	47.12	68.86	40.14
Zone 2 (West)								
EDTA extractable (mg/kg)	1.54	0.21	5.5	2.31	6	2.4	0.36	5.57
F1%	10.67	36.9	7.22	0.6	2.1	1.83	31.81	2.27
F2%	23.04	25.88	7.84	2.94	1.34	11.7	16.5	15.94
F3%	23.93	17.65	10.83	35.72	8.41	20.54	24.14	9.65
F4%	42.36	19.57	74.11	60.74	88.15	65.93	27.55	72.14
Zone 3 (East)								
EDTA extractable (mg/kg)	2.12	0.06	14.87	4.3	16.78	1.3	0.03	24.14
F1%	15.4	33.39	11.24	1.86	3.28	2.4	3.17	6.91
F2%	35.52	23.76	15.73	0.89	2.18	12.36	10.05	22.56
F3%	33.9	14.67	18.31	46.44	20.12	30.37	14.68	19.21
F4%	15.18	28.18	54.72	50.81	74.42	54.87	72.10	55.32
Zone 4 (South)								
EDTA extractable (mg/kg)	0.95	0.04	5.9	3.74	7.3	1.2	0.07	11.2
F1%	9.28	27.13	8.55	1.5	1.25	0.95	18.89	2.12
F2%	26.54	23.18	6.10	1.1	2	13.85	7.48	16.54
F3%	15.54	11.28	16.41	48.20	17.48	24.24	15.51	10.45
F4%	48.64	38.41	68.94	49.2	79.27	60.96	58.12	70.89

geogenic source. This is in accordance with Kabata-Pendias and Mukherjee [58].

Bioavailability and chemical partitioning of elements

Table 3 shows the elemental contents extracted by EDTA, and chemical partitioning of the four fractions (F1-exchangable fraction; F2-reducible fraction; F3-oxidizable fraction; and F4-residual fraction) obtained by sequential extraction. The mean EDTA extractable concentrations of elements are 26.73%, 67.30%, 7.12%, 27.70%, 6.65%, 23.55%, 18.84%, 23.89% of the mean total concentrations for As, Cd, Cr, Cu, Ni, Pb, Se and Zn, respectively. The highest bioavailability for As, Cd, Cu, Pb and Zn were measured in Zone 1, while Se was more available in soils of Zone 2. Also, Ni and Cr showed their highest EDTA-extractable concentrations (16.78 and 14.87 mg/kg, respectively) in Zone 3. A considerable risk of toxic metals, particularly Cd, for crops cultivated in soils of the study area could be concluded from these results.

Sequential extraction analyses revealed that Ni and Cr were mostly present in residual fraction. These metals have also the lowest proportion in exchangeable fraction (F1), confirming the results of EDTA method, and shows their bounding to resistant components of the solid matrix. Selenium has also a same situation with an exception for Zone 2 (west of the Kermanshah Province) with some oil fields. Cd and As are dominant in the first two fractions, particularly in Zones 1 and 3, respectively, while their lowest proportions are found in residual fraction. Copper showed the highest proportion in oxidisable fraction (organic matter and sulfides) in all four Zones, followed by Pb and As. This could be due to association of these elements with fertilizers used in agricultural soils of Kermanshah Province, or their affinity to organic matter. Moreover, the mean proportion of elements in non-residual fractions (the sum of three first steps of sequential extraction) decreased as Cd > As > Cu > Se > Pb > Zn > Cr > Ni, which is an estimation for mobility and bioavailability of studied elements in agricultural soils of Kermanshah Province.

Spatial distribution, contamination level and ecological risk

Figure 2 shows the spatial distribution of studied HMs in agricultural soils of Kermanshah Province. The interpolation methods used, were selected based on geostatistical data for the values of regression function (RF) and root mean squares

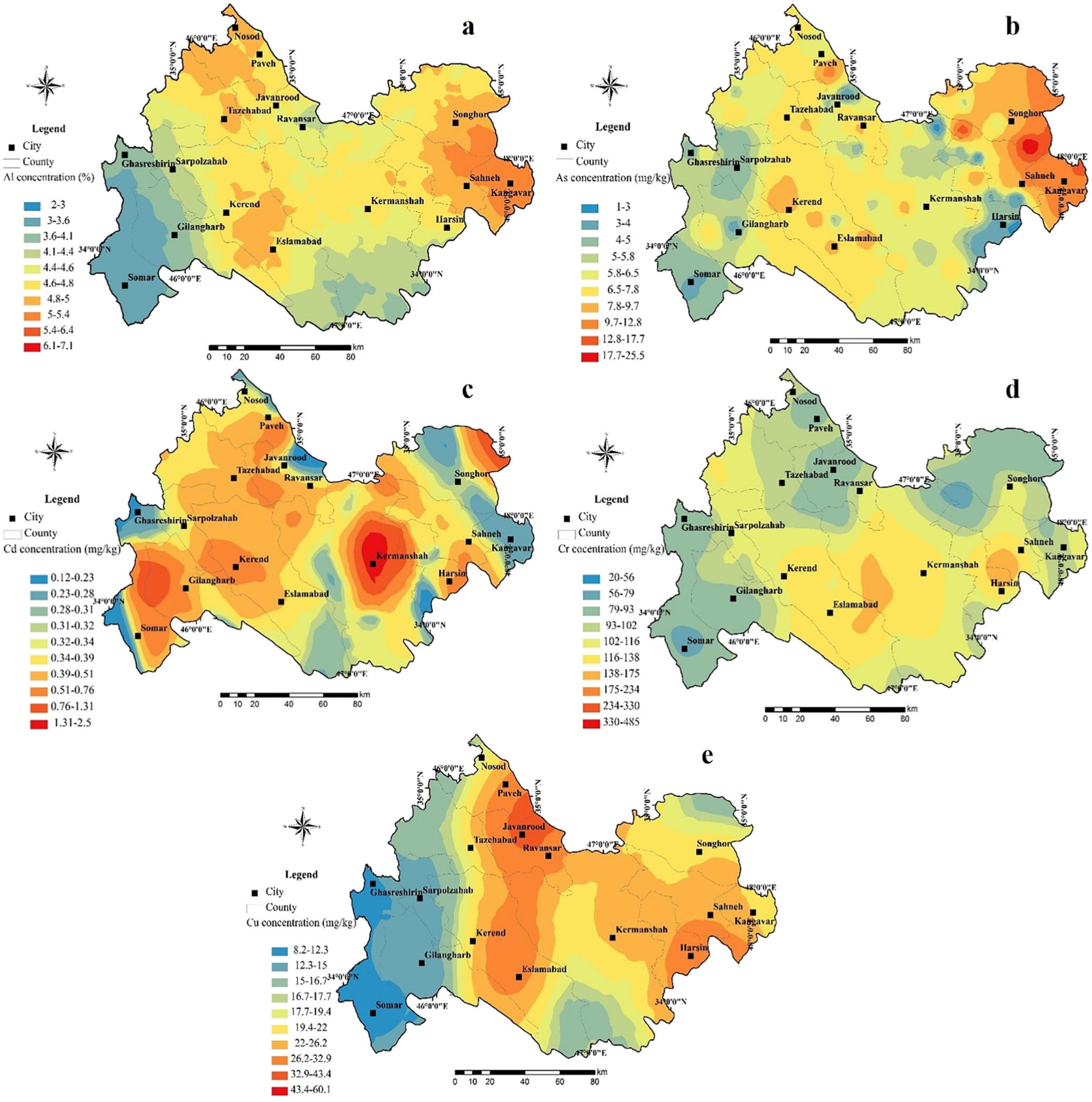

Fig. 2 Distribution maps of; **a**) Al, **b**) As, **c**) Cd, **d**) Cr, **e**) Cu, **f**) Ni, **g**) Pb, **h**) Se, **i**) Zn and **j**) SPI in agricultural soils of Kermanshah Province

of error (RMS). For this purpose, universal kriging, IDW, LPI, simple kriging, universal kriging, RBF, ordinary kriging, LPI and IDW methods were used for distribution of Al, As, Cd, Cr, Cu, Ni, Pb, Se and Zn, respectively. Four main spatial patterns were showed from the distribution maps. The spatial distribution maps for As, Cu, Pb and Zn, showed similar trends, with high contents in northeast and the center of the Province. Also, the spatial distributions of Se was obviously different, and high concentrations of this element was mainly found in the West. Cadmium showed its highest concentrations in agricultural soils of Kermanshah County and Western half of the Province. Moreover, relatively high concentrations of Cr and Ni were observed in Harsin and some parts of Kermanshah and Eslamabad Counties. As mentioned earlier, the main geological feature of these areas are basalt, gabbro and shale outcrops. Al showed no regular pattern in the soils samples.

Enrichment factor (EF) value at each station was calculated for selected HMs to get information about the levels of contamination, potential sources and anthropogenic disturbances. The results showed that the EF values of studied heavy metals followed the order of As > Cu > Pb > Se > Cd > Zn > Ni > Cr

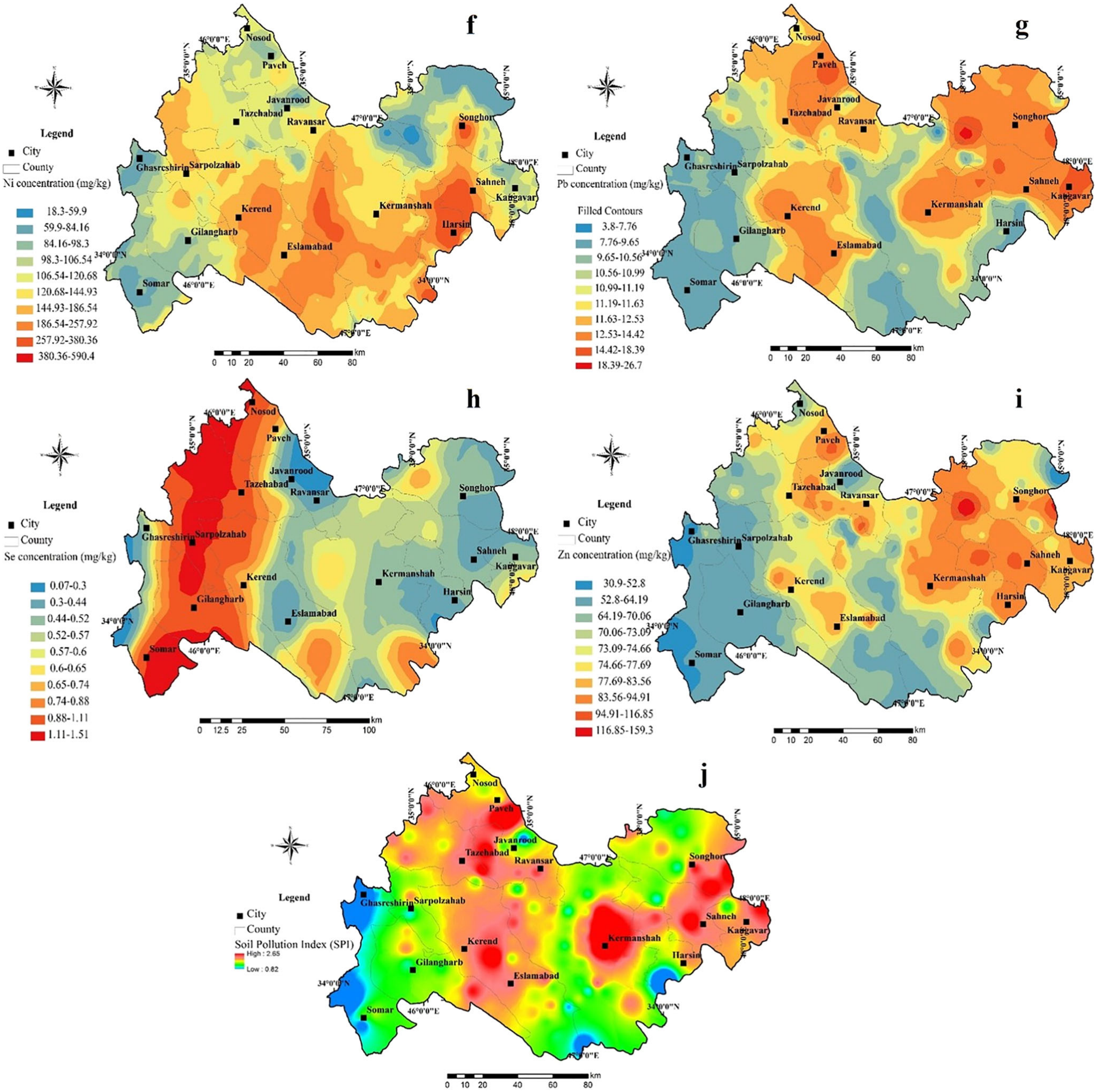

Fig. 2 continued.

(Fig. 3). Enrichment factors exceeding 1.0 suggest an anthropogenic source, and those lower than unity suggest a possible depletion of metals [59, 60]. The mean EF values of all the studied HMs (except Cr with mean EF of 0.89), were between 1 and 3, indicating minor enrichment. However, high EF values were calculated in some sampling stations. Cadmium showed moderate, and moderate to severe enrichment in three and four sampling stations, respectively. Also, moderate and severe enrichment were observed for Cu in four and one samples, respectively. Nickel, selenium and arsenic were also moderately enriched in five, nine and three agricultural soil sampling sites, respectively. Moreover, As and Se were moderately to severely enriched in one sample. Matching the results with sequential extraction and bioavailability tests, indicates that Ni enrichment is mostly relevance to outcrops of mafic and ultramafic rocks in some parts of the study area.

The enriched HMs in soils (As, Cd, Cu, Ni, Pb, Se and Zn) were used to calculated soil pollution index (SPI), and representation of the overall level of metals enrichment in soil (Fig. 2). In general, the SPI ranged from 0.82 to 2.65, with an average of 1.44. The values greater than 1 reveals soil pollution by selected elements. The relatively highly enriched

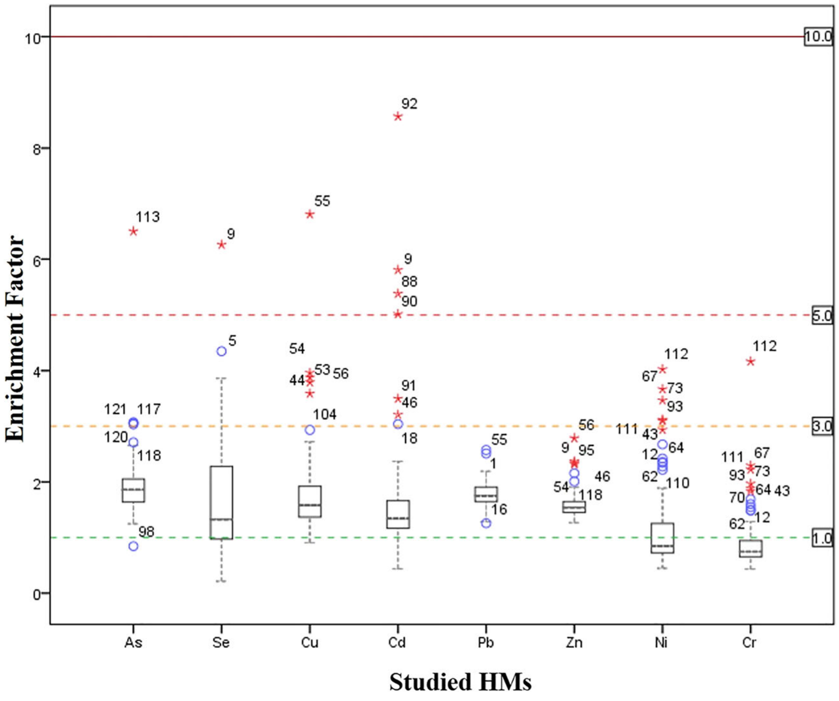

Fig. 3 Boxplot showing enrichment factor of HMs in agricultural soils of Kermanshah

areas were observed nearby Kermanshah County and Eastern parts of the Province. The main industries including oil refinery, petrochemical complex, chemical industries, Biseton power plant, and cement factory are located in this parts of the study area and may impact the agricultural soils. Also, the main transit road of Kermanshah, which connect the Province to Iran's Capital (Tehran) passes from this area. The soils of the Central and Northwestern parts (Eslamabad, Ravansar, Javanrood and Paveh Counties) were also enriched by HMs probably due to unmanaged use of pesticides, chemical fertilizers and sewage sludge in farm lands. These Counties along with Kermanshah are the main consumers of pesticides and fertilizers in the whole Province [24].

The potential ecological risk index (PER) of HMs and ecological risk index (E) of individual metals were calculated using the concentrations of seven studied metals (As, Cd, Cr, Cu, Ni, Pb and Zn). Figures 4 and 5 show the calculated E values of each metal, and distribution map of PER of the

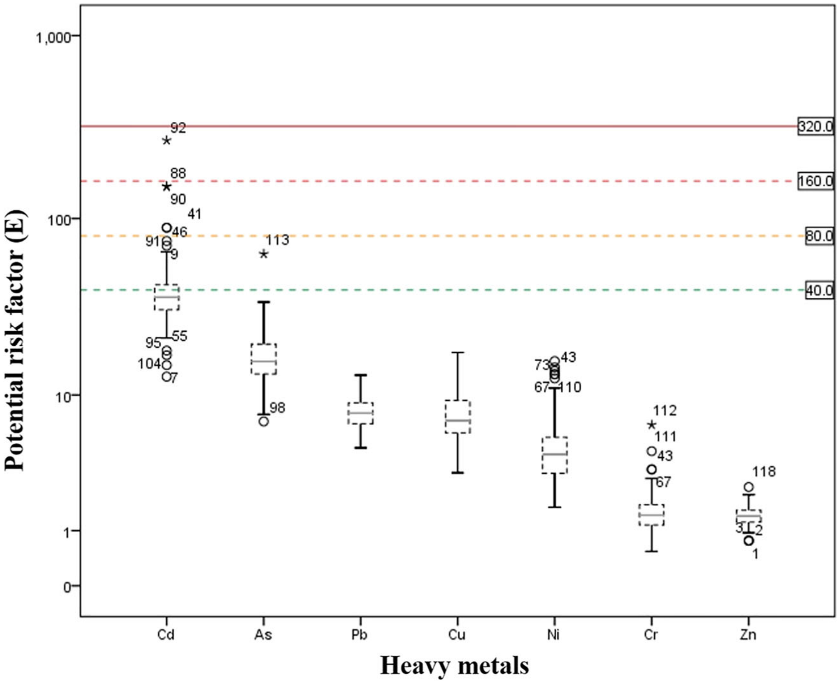

Fig. 4 Boxplot showing potential risk factors of individual HMs in Kermanshah agricultural soils

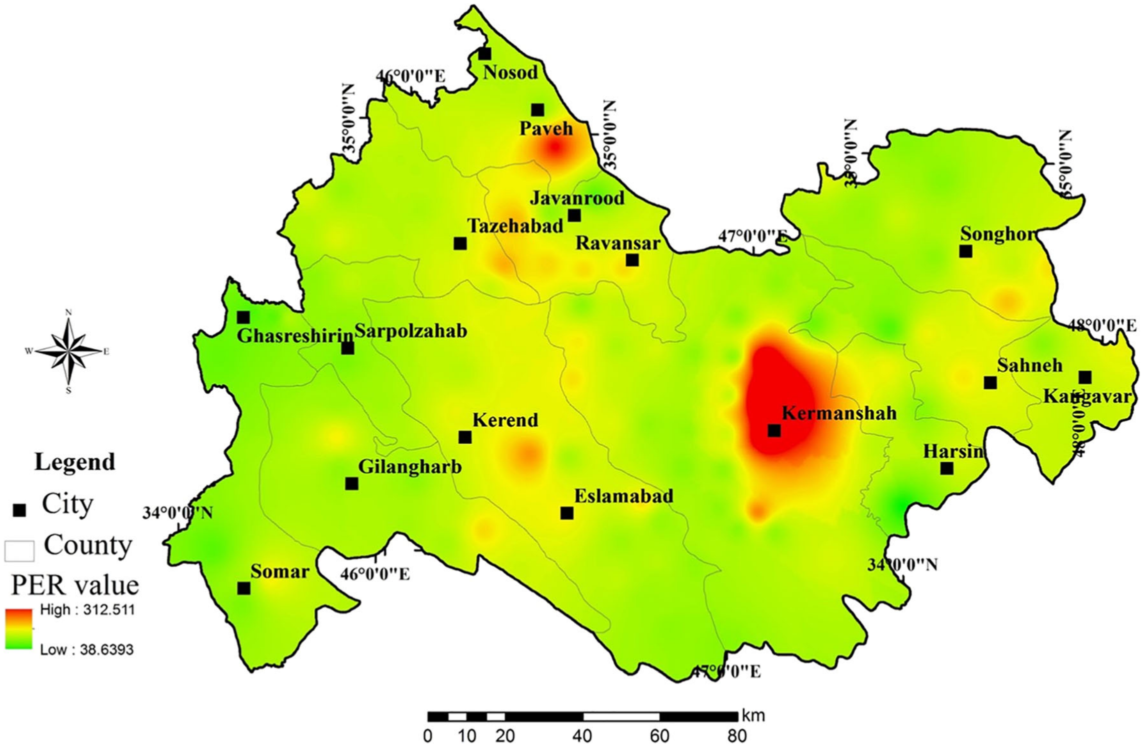

Fig. 5 Distribution map of PER of HMs in Kermanshah agricultural soils

seven studied elements in agricultural soils, respectively. Results showed that risk factors of all the studied HMs, except As (for one sample) and Cd, in all sampling stations bear low E value (below 40). Among the studied metals, Cd showed the highest E value due to its contaminations in some sampling stations and its high toxic-response factor. Also, based on PER values, except four samples with moderate potential ecological risk, agricultural soils were classified as low potential ecological risk (PER<150). The highest potential ecological risk were observed in Kermanshah (the center of the Province) and Paveh Counties. However, due to high bioavailability of some studied elements, particularly Cd and As, the actual and precise risk could be much higher.

Health risk assessment

Calculated hazard quotients and hazard index of different exposure pathways (for Cd, Cr, Cu, Ni, Pb and Zn), and cancer risk (for As, Cd, Cr, Ni and Pb) are presented in Table 4. Results showed that the highest HQ levels among the three exposure pathways belongs to the oral route of soil for both adults and children, except for Cd and Cr with highest HQ values for dermal exposure in children. Also, children showed higher values of both HQ and hazard index for all the studied metals than adults. However, except for Cr in children, the HI values of HMs were below unity for both children and adults, revealing no significant non-carcinogenic health risk. Based on the results of cancer risk calculation, As, Cd and Pb do not represent a carcinogenic risk for adults ($CR < 1 \times 10^{-6}$), but Cr and Ni represent a tolerable risk ($1 \times 10^{-6} < CR < 1 \times 10^{-4}$). On the other hand, for children, high risk of developing cancer is observed regarding Cr and Ni ($CR > 1 \times 10^{-4}$), and tolerable risk values of As, Cd and Pb are measured. It should be noted that based on sequential extraction and bioavailability tests, Cr and Ni had the lowest mobility in soils of the study area, and calculated CR and HQ values using total metals concentrations may have some uncertainties. Also, Cr valence state will directly affects its toxicity, and further detailed investigations are required.

There are some uncertainties for risk assessment in this study. For instance, Cr toxicity is dependent on its valence state, so that Cr (VI) is considered as carcinogen while Cr (III) is an essential factor for human health, but only the total content of Cr has been determined in this study. On the other hand, trace elements' interaction in the exposure process was not considered, nevertheless, this factor may change the results. Another source of uncertainty could be the differences in gender, age and body weight of individuals in an age group.

Statistical analysis

The multivariate analysis was performed to better identify the natural or anthropogenic sources of HMs, because the agricultural soils of Kermanshah Province showed enrichment in some metals compared to local baseline. Prior to principal component analysis (PCA), the data was log-transformed,

Table 4 Health risk of heavy metals in the studied soil samples

Element	As	Cd	Cr	Cu	Ni	Pb	Zn
HM (95% UCL)	7.41	0.45	125.64	22.23	163.41	11.89	75.23
Oral RfD	–	1.00E-03 [66]	3.00E-03 [66]	4.00E-02 [66]	2.00E-02 [69]	3.50E-02 [66]	3.00E-01 [70]
Dermal RfD	–	5.00E-05 [66]	6.00E-05 [66]	1.20E-02 [66]	5.40E-03 [69]	5.30E-04 [66]	6.00E-02 [70]
Inh. RfD	–	1.00E-03 [67]	2.86E-05 [66]	4.02E-02 [66]	2.06E-02 [69]	3.52E-03 [66]	3.00E-01 [67]
SF	1.5 [61]	15 [68]	5.00E-01 [65]	–	8.40E-01 [69]	2.80E-01 [66]	–
Children							
$\mathrm{HQ}_{\mathrm{ing}}$	–	2.32E-03	2.16E-01	2.87E-03	4.21E-02	2.04E-02	1.29E-03
$\mathrm{HQ}_{\mathrm{derm}}$	–	7.51E-03	1.75E+00	1.55E-03	2.53E-02	1.87E-02	1.05E-03
$\mathrm{HQ}_{\mathrm{inh}}$	–	1.30E-07	1.27E-03	1.59E-07	2.29E-06	9.73E-07	7.22E-08
HI	–	9.83E-03	1.97E+00	4.41E-03	6.74E-02	3.92E-02	2.34E-03
CR	6.66E-05	4.04E-05	3.76E-04	–	8.22E-04	1.99E-05	–
Adults							
$\mathrm{HQ}_{\mathrm{ing}}$	–	2.70E-04	2.51E-02	3.33E-04	4.90E-03	2.04E-03	1.50E-04
$\mathrm{HQ}_{\mathrm{derm}}$	–	2.98E-05	6.94E-03	6.14E-06	1.00E-04	7.44E-05	1.25E-05
$\mathrm{HQ}_{\mathrm{inh}}$	–	7.93E-08	7.74E-04	9.75E-08	1.40E-06	5.95E-07	4.42E-08
HI	–	3.00E-04	3.28E-02	3.39E-04	5.00E-03	2.11E-03	1.63E-04
CR	6.7E-06	4.07E-06	3.79E-05	–	8.27E-05	2.01E-06	–

since the Kolmogrov-Smisnov normality test ($p > 0.01$) revealed that the data is not normally distributed. Three principal components with eigenvalues higher than 1 (before and after Varimax rotation) were extracted (Table 5). PCA resulted in a reduction of the initial dimension of the dataset to the components, explaining a 79.81% of the data variation. Factor 1, explaining 39.63% of the total variance, had a high loading of As, Cu, Pb, Zn and somewhat Cd, and can be considered as an anthropogenic component. According to the results of EF calculations, industries including oil refinery, petrochemical complex, chemical industries, Biseton power plant, and cement factory, and also application of chemical fertilizers, sewage sludge and pesticides, are the main anthropogenic sources of the HMs in the area. Factor 2 loadings on Al, Cr and Ni, and explains 22.88% of the total variance, which can be considered as natural source and poorly affected by anthropogenic sources. This is in accordance with the results of enrichment factor calculation, and bioavailability and sequential extraction analyses. The third factor includes Se and explained 17.3% of the total variance. Although based on enrichment factor, like the elements of component 2, Se was enriched and therefore have an anthropogenic source, this element has placed in a separate component. This could be due to different sources in the area. On the other hand, the highest Se concentrations were observed in Western parts of the Province, while the highest Pb and Zn concentrations were observed in Eastern parts.

Table 5 Varimax-rotated factor model for soil samples

	Component		
	1	2	3
Cd	0.58	0.16	0.35
Cr	0.15	0.97	0.03
Cu	0.53	0.20	−0.66
Ni	0.04	0.99	0.00
Se	−0.16	0.06	0.92
Zn	0.87	0.14	−0.12
Al	0.21	0.91	−0.10
As	0.75	0.03	−0.06
Pb	0.93	−0.08	−0.21

Due to non-normal distribution of the data, Kruskal-Wallis H test was used to compare HMs concentrations in agricultural soils of 16 different Counties in the study area (Table 6). In this test, p value below 0.05, indicates statistically significant difference between the medians of the groups. Results revealed that median concentrations of all the studied elements, except Al, are significantly different between the investigated Counties ($p < 0.05$). Various anthropogenic sources for HMs placed in components one and three of PCA (and Ni in five samples), along with some gabbro and basalt outcrops as geogenic source for Cr and Ni [58] could be the main reasons for this statistically significant difference between soil elemental concentrations in different Counties of Kermanshah Province.

Table 6 Kruskal-Wallis H test to compare HMs concentrations in soils of different Counties

	Cd	Cr	Cu	Ni	Se	Zn	Al	As	Pb
Chi-Square	34.18	39.13	64.31	45.15	56.28	36.71	29.84	39.73	39.02
df	15.00	15.00	15.00	15.00	15.00	15.00	15.00	15.00	15.00
Asymp. Sig.	0.003	0.001	0.00	0.00	0.00	0.001	0.073	0.00	0.001

Conclusions

In the present study, the pollution status, bioavailability and partitioning, spatial distribution, potential sources, and health and ecological risk of selected heavy metals in agricultural soils of Kermanshah Province were investigated. The results revealed the role of various industries and application of fertilizers/pesticides (for As, Cd, Cu, Pb, Se and Zn), and geology of the study area (particularly for Cr and Ni) in different distribution patterns, enrichment levels and health risks. Kermanshah County, showed the highest soil HMs enrichment with highest bioavailability, and potential ecological risk in the Province. Among the studied elements, the highest ecological risk factor was calculated for cadmium, which regarding its high mobility and bioavailability in soils of the study area, may cause severe pollution of food crops. Also, high proportion of Cd, As and Cu in the first three fractions of sequential extraction, shows their high bioavailability and potential risk in case of changing soil physicochemical parameters. However, Considering low concentrations of Zn, as an essential element, and it's mostly presence in residual fraction, except in Zone 1, deficiency assessment of this metal and other micronutrients in soil and food crops of Kermanshah Province is recommended. On the other hand, exposure to soils of the study area may cause cancer and other non-cancerous disorders, mainly by dermal contact and digestion of contaminated soil. Regarding these results, a better management of contamination sources in the area is required to minimize the contamination of cultivated crops and preventing residents of the area from being exposed to contaminated soil.

Acknowledgments The authors wish to express their gratitude to the Iran Department of Environment. Thanks are extended to the Research Committee of Kharazmi University for Logistic Help.

Compliance with ethical standards

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

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