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Multivariate statistical analysis of heavy metals contamination in atmospheric dust of Kermanshah province, western Iran, during the spring and summer 2013

Shahab Ahmadi Doabi[a,*], Majid Afyuni[a], Mahin Karami[b]

[a] Department of Soil Science, College of Agriculture, Isfahan University of Technology, Isfahan, 84156-83111, Iran
[b] Department of Soil Science, Agricultural Engineering Campus, Razi University, Kermanshah, 6715685438, Iran

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ABSTRACT

Atmospheric dust is known to contain heavy metals in different levels that can cause environmental pollution. In west of Iran, despite of the large number of dusty days per year, heavy metals content of the atmospheric dust has not been previously investigated. A total of 98 samples of atmospheric dust were collected during the spring (T1) and summer (T2) of 2013 from urban and suburban locations in Kermanshah province, and analyzed for Zn, Cu, Ni, Cr, Mn, and Fe total concentrations. Correlation, principal component and cluster analyses suggested probable natural and anthropogenic sources of the metals in the dust. The contamination levels of heavy metals were assessed on the basis of enrichment factor (EF), geo-accumulation index (I_{geo}), and ecological risk (RI). Comparing with the background values of world soils, elevated metal concentrations were found in dust samples, except for Mn and Fe. Manganese and Fe were mainly of natural origin with traces of anthropogenic influences; while Zn and Cu were mainly from traffic sources and probably were partly from industrial sources; Ni and Cr mainly resulted from industrial activities and probably in part from traffic sources. The analysis of EF revealed moderate enrichments for Mn and Cr, and significant enrichments for Zn, Cu, and Ni in T1 and T2, and similarly in urban and suburban areas. Based on I_{geo} index, the levels of Mn and Fe were classified as uncontaminated, while Zn, Cu, Ni, and Cr levels were evaluated to have moderate contamination in T1 and T2, and similarly in urban and suburban areas. The values of RI showed a low level of heavy metals pollution.

1. Introduction

Of the three materials, soil, sediment and dust, which originate primarily from the earth's crust, dust is the most pervasive and important factor affecting human health and well-being (Banerjee, 2003; Yongming et al., 2006). In urban areas, dusts are indicators of environmental pollution due to atmospheric deposition (Li et al., 2001; Yongming et al., 2006; Al-Khashman, 2004).

As an important group of dust constituents, heavy metals are natural components of the Earth's crust (Tokalıoğlu and Kartal, 2006; Al-Khashman, 2004). They do not degrade or destroy and can remain in soil and dust over long periods of time (Tokalıoğlu and Kartal, 2006; Saeedi et al., 2012). Elevated levels of heavy metals are ubiquitous in urban settings as the result of a wide range of human activities, especially from industrial sources (Duzgoren-Aydin et al., 2006). As a result, the adverse effects of poor environmental conditions on human health are most evident in urban environments, particularly in developing countries where urbanization, industrialization and rapid population growth are taking place on an unprecedented scale (Atiemo et al., 2011). Moreover, heavy metals in street dusts could easily enter human bodies through dust ingestion, inhalation and dermal contact under dynamic conditions such as wind, traffic and other human activities (Ewers, 1991; Atiemo et al., 2011; Wei et al., 2010a, 2010b; Wei et al., 2015). Numerous studies have shown that heavy metals are persistent and widely dispersed in the environment, interact with different natural components, and pose threats to human health and the environment (Vives et al., 2006; Park and Dam, 2010; Aničić et al., 2011; Pavlík et al., 2012). The adverse effects of heavy metals in road dust include respiratory system disorders, nervous system interruptions, endocrine system malfunction, immune system suppression and the risk of cancer in later life (Ferreira-Baptista and De Miguel, 2005).

Heavy metals may originate from various types of anthropogenic sources such as petroleum, diesel and coal combustion, as well as industrial activities (Loredo et al., 2003; Manasreh, 2010) and natural geochemical processes such as weathering (Saeedi et al., 2012). In urban areas, the metals found in street dust may come from many

* Corresponding author.
E-mail address: shahab.ahmadi@ag.iut.ac.ir (S.A. Doabi).

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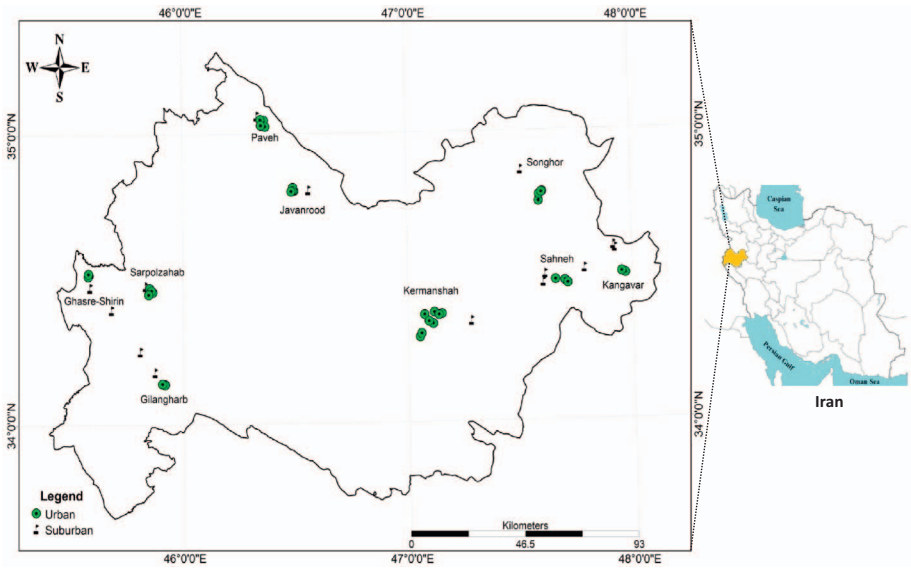

Fig. 1. Study area and sampling sites in Kermanshah province.

different sources, including vehicle emissions, industrial discharges, weathered materials and various human activities (Al-Khashman, 2004; Gibson and Farmer, 1986; Harrison et al., 1981; Li et al., 2001; Sezgin et al., 2004; Thornton, 1991). Main anthropogenic sources of heavy metals exist in various industrial point sources, e.g. present and former mining activities, foundries, smelters and diffuse sources such as piping, constituents of products, combustion by-products, traffic, industrial and human activities (Al-Khashman, 2004). Three main factors known to influence the levels of heavy metals in dust samples which have been reported are traffic, industry and weathered materials, particularly house and street dust (Arslan, 2001). The sources of Cu, Zn, and Pb in urban areas are mainly traced to vehicle exhaust emissions and the wear products from road transportation (Harrison et al., 2003). Additionally, exhaust gases and fossil fuel combustion have been claimed as sources of Ni and Zn (Espinosa et al., 2001; Mugica et al., 2002; Wang et al., 2005), while Fe and Mn have been traced to steel industrial processes and the Earth's crust (Mugica et al., 2002; Wang et al., 2005). The source of Ni and Cr in street dust is believed to be due to corrosion of vehicular parts (Lu et al., 2009; Akhter and Madany, 1993; Fergusson and Kim, 1991) and chrome plating of some motor vehicle parts (Al-Shayeb and Seawardt, 2001). Industrial metallurgic processes have also been reported to increase As, Cd, Cu, Ni, and Zn concentrations in the environment (Wang et al., 2005; Park and Dam, 2010).

The rapid growth of industry, population and vehicle exerts a heavy pressure on the urban environment. Though there are many studies of heavy metal contamination of urban dusts in developed countries, little attention has been paid to this matter in developing countries (Banerjee, 2003; Awan et al., 2011). The atmospheric dust pollution with heavy metals has not been previously investigated in western Iran. In recent years, the region has been experiencing dust pollution frequently some probably coming from the western countries in the Middle East. On the other hand, limited information is available on dust heavy metal content in the Middle East region. Considering identification of metal sources and assessing the extent of heavy metals pollution present in atmospheric dust is important for establishing pollution control strategies (Loska and Wiechuła, 2003; Wei and Yang, 2010; Ghrefat et al., 2011). The main purposes of the present study were to: (a) determine total concentrations of Zn, Cu, Ni, Cr, Mn, and Fe in atmospheric dusts from different areas of Kermanshah province (b) identify their natural or anthropogenic origin by using correlation analysis, principal component analysis (PCA) and cluster analysis (CA), and (c) assess the levels of contamination based on the enrichment factor (EF), geo-accumulation index (I_{geo}), and ecological risk (RI). The results can provide a baseline for use in future environmental impact assessments and to guide pollution mitigation targets in the region.

2. Materials and methods

2.1. Study area

This study was conducted in 9 townships in Kermanshah province, located in the west part of Iran (45°24′-48°7′ E, 33°40′-35°18′ N) at an average altitude of 1200 m above the sea level. The province covers an area of 25,008 km^2 and is the twelfth most populated province in Iran with a population of about 1,945,227 inhabitants (Statistical Center of Iran, 2011) with average density of 78 Persons/km^2. The climate in this area is arid and semi-arid with annual mean temperature and precipitation of 16 °C and 450 mm, respectively. The prevailing wind directions are west to east with northwest and southwest fluctuations (IRIMO, 2013).

Gaseous wastes in the form of automobile exhaust, chemicals factories emissions, different kinds of industries (including an oil refinery and a petrochemical factory) and primitive forms of heating, as well as dust input from Iraq, the neighboring country, are the major sources of air pollution in the province. In addition to high population growth, the rate of urbanization has also accelerated, and is now one of the highest in Iran.

2.2. Atmospheric dust sampling

A total of 98 dust samples were collected in a temporal range from spring to summer, each sample lasting 87 days from different urban (35 site) and suburban (14 site) locations in Kermanshah, Songhor, Gilangharb, Ghasre-Shirin, Sahneh, Sarpolzahab, Kangavar, Paveh, and Javanrood cities. The sampling sites were roughly distributed over the urban and suburban areas (Fig. 1). Kermanshah province meteorological organization statistics showed dusty days occur mainly in spring and summer seasons.

The sampling sites were selected based on the following criteria. They were not shaded by trees or buildings, were easily accessible and secure against interference by animals, humans and upwind obstructions. Accordingly, dust collectors (passive samplers) were installed on the roof of buildings about 3–4 m above the ground level. Each collection tray consisted of a circular plastic surface (320 mm in diameter, 120 mm depth) fixed on holders with 33 cm height and were covered with a 2 mm PVC mesh on top to form a rough area for trapping saltant particles.

Dry sample weight, surface area exposed by each trap (0.08 m^2),

and the period of dust collection were used to calculate dust deposition rates. Dust samples were collected carefully with distilled water and stored in sealed polyethylene bags, labeled and then transported to the laboratory. Total of 98 dust samples for two seasons were oven dried for 24 h at temperature 105 °C to a constant mass and then their weight was recorded.

2.3. Chemical analysis

Dust samples were sieved through a 1 mm mesh. An accurately weighed 0.5 g of dust was placed in a test tube, 10 ml of a 3:1 concentrated HCl/HNO_3 mixture was added to each test tube, and the mixture was left at room temperature overnight. Each test tube was covered with an air condenser and refluxed gently at 80 °C for 2 h. After cooling, the solution was filtered through a moistened Whatman 42 filter paper and diluted to 50 ml volume with distilled water (Karimi et al., 2009; Sparks et al., 1996). The final solutions were analyzed for their Zn, Cu, Ni, Cr, Mn, and Fe concentrations using an Atomic Absorption Spectrophotometer (AAS: model Perkin Elmer 3030, USA). The detection limit values for all studied elements were between 0.01 and 0.02 ppm, with wavelengths of 213.9, 324.8, 232, 357.9, 279.5 and 248.3 for Zn, Cu, Ni, Cr, Mn and Fe, respectively. The standard solution concentrations were 0.5, 1.5, 3, for Zn and Cr, 2, 5, 15, 30 for Fe, 0.5, 1.5, 3 or 2, 6, 12 for Cu and Mn, and 0.5, 1.5, 3 or 1, 3, 6 for Ni. For Cu, Ni and Mn one of the two groups of standard solutions were used based on the sample content of the element. In fact, first group were applied for samples with less element content and second group for samples with greater concentration of the element.

2.4. Statistical analysis

To evaluate the obtained results, correlation analysis, principal component analysis (PCA) and cluster analysis (CA) were used. Pearson's correlation coefficients were used to identify the relationships between the six heavy metals. PCA and CA are the most common multivariate statistical methods used in environmental studies (Yongming et al., 2006; Tahri et al., 2005).

PCA, is widely used to extract a smaller number of independent factors (principal components) among available data for analyzing variables relationships (Tokalıoğlu and Kartal, 2006; Yongming et al., 2006). PCA can reduce the number of correlated variables to a smaller set of orthogonal factors, making it easier to interpret a given multidimensional system by displaying the correlations among the original variables. Principal components are those whose eigenvalues exceed 1 (Kaiser, 1960). PCA have been widely applied to various environmental media, to identify pollution sources and to apportion natural versus anthropogenic contributions (Lu et al., 2010). The components of the PCA were transformed using a varimax rotation with Kaiser Normalization after the analysis (Yongming et al., 2006).

CA classifies a set of observations into two or more mutually exclusive unknown groups based on a combination of internal variables (Lu et al., 2010). Cluster analysis has been applied to identify different geochemical groups by clustering the samples with similar heavy metal content (Yongming et al., 2006; Lu et al., 2010). A dendrogram is the most commonly used method of summarizing hierarchical clustering (Lu et al., 2010). In this study, CA was performed according to Ward's method (Yongming et al., 2006; Tokalıoğlu and Kartal, 2006). The results were displayed as a dendrogram created using hierarchical clustering, and values of the distances between clusters (the Euclidean distance) were presented.

In the current study, PCA and CA were used to elucidate the latent relationships between variables and/or samples, and for investigating pollutants (variables) sources. In our study, statistical analyses were performed using SPSS package version 16.0 for Windows. Since multivariate statistical analysis is sensitive to outliers and non-normality, Johnson-transformation method (Unbounded system distribution for Zn, Cu, Ni and Cr in T1, and Cu, Ni and Cr in T2, and Bounded system distribution for Zn and Mn in T2) was performed for data normalization except for Mn and Fe in T1 and Fe in T2 (those data were normal originally and did not need any transformation) and a Kolmogorov–Smirnov (K–S) test was used for checking data normality.

2.5. Pollution level assessment methods

2.5.1. Enrichment factor

The enrichment factor (EF) method described by Sutherland (2000) was used to evaluate the potential impact of the dust samples. The EF of each element, which is a normalization of an element of interest against a reference one, was calculated using the following equation:

$$EF = (C_x/C_{ref})_{sample}/(C_x/C_{ref})_{background} \quad (1)$$

to determine the metal enrichment in dusts and probable natural or anthropogenic sources (Wei et al., 2010b; Manasreh, 2010). Here (C_x/C_{ref}) is the ratio of concentrations between a heavy metal and a reference metal in the sample and background (Alloway, 2010). We selected Iron (Fe) as the reference metal based on correlation coefficient analysis and multivariate statistical analyses from metals that are neither likely to be affected by anthropogenic activities nor correlated with heavy metal pollutants (Saeedi et al., 2012; Keshavarzi et al., 2015). Iron is commonly used, as reference element (Atiemo et al., 2011; Yongming et al., 2006, Tasdemir and Kural, 2005; Turner and Simmonds, 2006). The EF was split into five classes as follows: $EF < 2$, class 1, deficiency to minimal enrichment; $2 \leq EF > 5$, class 2, moderate enrichment; $5 \leq EF > 20$, class 3, significant enrichment; $20 \leq EF > 40$, class 4, very high enrichment; and $EF \geq 40$, class 5, extremely high enrichment (Yongming et al., 2006; Kartal et al., 2006).

2.5.2. Geo-accumulation index (I_{geo})

The I_{geo}, introduced by Müller (1969), is defined by the following equation:

$$I_{geo} = \log_2[C_i/(1.5C_{ri})] \quad (2)$$

where C_i represents the measured concentration of the element i in the sample and C_{ri} is the geochemical background value or reference value of the element i. In this study, C_{ri} is the background content of element i in world soils (Alloway, 2010). The constant 1.5 is the background matrix correction factor due to lithological variability. The following classification is given for geo-accumulation index (Huu et al., 2010; Müller, 1981): $I_{geo} \leq 0$, class 0, practically unpolluted; $0 < I_{geo} \leq 1$, class 1, unpolluted to moderately polluted; $1 < I_{geo} \leq 2$, class 2, moderately polluted; $2 < I_{geo} \leq 3$, class 3, moderately to strongly polluted; $3 < I_{geo} \leq 4$, class 4, strongly polluted; $4 < I_{geo} \leq 5$, class 5, strongly to extremely polluted; and $I_{geo} > 5$, class 6, extremely polluted.

2.5.3. Ecological risk

The method of determining ecological risk of heavy metals originally introduced by Hakanson (1980), has recently been used in dust contamination studies (Tang et al., 2013). Hence, the potential ecological risk index (PERI) was calculated to assess the degree of metal pollution in Kermanshah atmospheric dust as follows:

$$RI = \sum_{i=1}^{m} E_r \quad (3)$$

$$E_r = T_r \times C_f \quad (4)$$

$$C_f = \frac{C_s}{C_n} \quad (5)$$

where C_s and C_n are heavy metal concentrations in sample and background (Alloway, 2010), respectively, E_r is the ecological risk of each element and RI shows the ecological risk of multiple elements.

Table 1
Heavy metals concentrations (mg kg^{-1}) in atmospheric dust collected from Kermanshah province during the spring (T1, $n = 49$) and summer (T2, $n = 49$) 2013, and in background soils of world.

Element	Sampling time	Minimum	Maximum	Mean	Median	S.D.	CV	Skewness	Urban	Suburban	Background-values (Alloway, 2010)
Zn	T1	88	490	182.29	146	91.49	0.50	1.99	186.75	171.14	62
	T2	132	700	238.29	191	126.07	0.53	1.96	242.82	226.99	
Cu	T1	35	110	48.59	45	11.61	0.24	3.27	50.65	43.43	14
	T2	24	256	46.67	37.58	34.66	0.74	4.98	47.37	44.92	
Ni	T1	69	238	115.33	99.20	36.28	0.32	1.32	118.30	107.91	18
	T2	60	245	123.72	101	51.17	0.41	0.88	131.59	104.06	
Cr	T1	44	119	73.87	68	17.84	0.24	0.99	76.28	67.86	42
	T2	44	147	73.60	62.50	25.62	0.35	1.35	77.46	63.94	
Mn	T1	345	495	428.07	424	32.63	0.08	− 0.05	425.89	433.50	418
	T2	400	695	495.16	482	56.60	0.11	0.96	494.31	497.27	
Fe	T1	13,150	33,150	23,161.22	23,200	3285.29	0.14	0.30	22,958.57	23,667.86	47,000
	T2	20,750	35,562	28,703.94	28,691.70	2956.44	0.10	− 0.15	28,617.10	28,921.05	

Hakanson (1980) defined T_r as a "toxic-response factor" for a given substance and demonstrated this value for Cu, Ni, Cr, Zn, and Mn to be 5, 5, 2, 1, and 1, respectively. The following terminologies are used to describe risk levels: $E_r < 40$, low potential ecological risk; $40 \leq E_r < 80$, moderate potential ecological risk; $80 \leq E_r < 160$, considerable potential ecological risk; $160 \leq E_r < 320$, high potential ecological risk; and $E_r \geq 320$, very high ecological risk; $RI < 150$, low ecological risk; $150 \leq RI < 300$, moderate ecological risk; $300 \leq RI < 600$, considerable ecological risk; and $RI \geq 600$, very high ecological risk.

3. Results and discussion

3.1. Heavy metal concentrations

The descriptive statistic results of heavy metal concentrations, as well as background values of world soils (Alloway, 2010), are presented in Table 1. Except for Mn and Fe, all heavy metals concentrations of atmospheric dust samples were greater than concentrations in the background soils of the considered localities of the world. The average concentrations of Zn, Cu, Ni, and Cr in urban dust samples were more than suburban samples for both seasons (T1 and T2), in agreement with the results of Tang et al. (2013). However, concentrations of Mn and Fe were detected in dust samples from suburban more than urban areas (Table 1).

Large standard deviations were found for all the heavy metals. This indicates a wide variation of concentrations in atmospheric dust samples. Skewness values of heavy metals are positive except for MnT1 and FeT2, showing that mean concentrations are higher than their median concentrations. The skewness values for CuT1 and CuT2 were 3.27 and 4.98 (maximum value among all elements), which indicate the existence of highly contaminated spots. In addition, the high coefficient of variation (CV) (especially for ZnT1, ZnT2, and CuT2) indicates a high inhomogeneity of elements in dust samples of the present study (Table 1). The values of the mean concentrations of heavy metals in atmospheric dust of Kermanshah divided by the corresponding values of the background soils (from the considered localities of the world), decrease in the order of Ni > Cu ~ Zn > Cr > Mn > Fe (Tables 1 and 2).

Nickel concentration in Kermanshah dusts is significantly greater than for all other cities in the world (Table 2). In general, elevated Ni concentration in urban dusts can be attributed to vehicle instruments because of the engines using nickel gasoline as fuel (Al-Khashman, 2007). Chromium concentration in Kermanshah dust is more than for other cities, except for Nanjing and Baoji. The mean concentration of Mn in dust samples of Kermanshah is higher than those in Amman, Mutah, Tokat and Madrid, also close to its background value. On the other hand, the mean concentration of Fe in Kermanshah atmospheric dust is higher than for the other sampled cities, except for Tehran, Nanjing and background value.

3.2. Correlation coefficients analysis

Pearson's correlation coefficients of heavy metals in Kermanshah atmospheric dust from two sampling times are summarized in Table 3. Nickel correlated significantly positively with Cr (T1 = 0.82 and T2 = 0.54) at $P < 0.01$, such correlations were not observed with Mn or Fe in both sampling times. Another group is based on the strong correlation between Zn and Cu (T1 = 0.55 and T2 = 0.61, $P < 0.01$) that suggests strong association and a common source. Also, Zn and Cu

Table 2
Comparison of the heavy metals contents in atmospheric dust of Kermanshah province with reported values for other cities in previous studies (mg kg^{-1}).

Location	Zn	Cu	Ni	Cr	Mn	Fe	Reference
Kermanshah[a]	210.29	47.63	119.53	73.74	461.61	25,932.58	Present study
Shiraz (Iran)	403.5	136.3	77.5	67.2	438.5	20,255	Keshavarzi et al., 2015
Kurdistan (Iran)	–	61.3	–	–	497.7	14,003.6	Khuzestani and Souri, 2013
Arak (Iran)	9.3	45.1	28.5	45.9	–	–	Ghadimi et al., 2013
Tehran (Iran)	873.2	225.3	34.8	33.5	1214.5	47,935.7	Saeedi et al., 2012
Nanjing (China)	394	123	55.9	126	646	34,200	Hu et al., 2011
Baoji (China)	715.3	123.2	48.8	126.7	804.2	–	Lu et al. 2009 and 2010
Amman (Jordan)	401	249.6	16.3	18.3	144.6	5370.6	Jiries, 2003
Mutah (Jordan)	132	69	1.7	–	136	5362	Manasreh, 2010
Tokat (Turkey)	63	29	65	30	285	–	Kurt-Karakus, 2012
Ottawa (Canada)	184	188	19	59	534	25,660	Rasmussen et al., 2001
Newcastle (UK)	421	132	26	–	–	–	Okorie et al., 2012
Madrid (Spain)	476	188	44	61	362	19,300	De Miguel et al., 1997

[a] Average for T1 and T2.

Table 3
Pearson's correlation coefficients matrix among the heavy metals concentrations in the atmospheric dust of Kermanshah province in times T1 and T2.

Sampling time	Element	Zn	Cu	Ni	Cr	Mn	Fe
T1	Zn	1					
	Cu	0.55[a]	1				
	Ni	0.55[a]	0.46[a]	1			
	Cr	0.60[a]	0.52[a]	0.82[a]	1		
	Mn	− 0.12	0.23	− 0.04	0.07	1	
	Fe	0.19	0.28	0.27	0.42[a]	0.52[a]	1
T2	Zn	1					
	Cu	0.61[a]	1				
	Ni	0.44[a]	0.46[a]	1			
	Cr	0.59[a]	0.61[a]	0.54[a]	1		
	Mn	0.39[a]	0.41[a]	0.05	0.44[a]	1	
	Fe	0.52[a]	0.57[a]	0.16	0.70[a]	0.70[a]	1

[a] Correlation is significant at the 0.01 level (two-tailed).

correlate well with other elements except for Mn and Fe in T1. At the T1 sampling time, Mn and Fe were only correlated to each other significantly positively at $P < 0.01$ (0.52), while in T2 in addition to the strong correlation between Mn and Fe (0.70, $P < 0.01$), a good correlation between these two metals with Zn, Cu, and Cr were observed.

3.3. Multivariate statistical analysis

3.3.1. Principal component analysis

The PCA was applied to identify possible sources of pollutants in atmospheric dust by applying varimax rotation with Kaiser normalization in times T1 and T2. Table 4 shows the results of the factor loadings with a varimax rotation, as well as the eigenvalues and communalities. Principal factors, extracted from the variables with eigenvalues > 1, were selected. As expected, three factors were acquired. Principal factors > 0.6 are bolded in each column. A three-dimensional plot of the PCA loadings is presented in Fig. 2, and the relationships among the six heavy metals are readily observed. As expected, three factors were obtained for T1 and T2, accounting for 84.85% and 84.68% of the total variance, respectively. At T1, the first factor explains 35.50% of the total variance and loads heavily on Ni and Cr, while at T2, it explains 34.07% of the total variance and loads on Mn and Fe. Factor 2, dominated by Mn and Fe at T1 and, Zn and Cu at T2, accounts for 26.08% and 26.27% of the total variance, respectively. Finally, factor 3 is dominated by Zn and Cu; accounting for 23.27% of the total variance at T1, and by Ni and Cr; accounting for 24.34% of the total variance at T2. The Ni loading (0.92) is as high as the Cr loading (0.65), which may imply quasi-independent behavior within the group. Fig. 2 (T2) shows that, Cr and Ni are separated by a large distance in the three-dimensional PCA loading plot, which may suggest that the two elements are poorly correlated and have different sources. These results indicate that factors 1 and 3 in T1 and 2 and 3 in T2 are likely originate from common anthropogenic sources, whereas, factors 2 in T1 and 1 in T2 might be from natural origins. The main anthropogenic sources in the region include traffic emissions, industrial activities and mining activities.

3.3.2. Cluster analysis

Before CA, the values were standardized using z-scores, the Euclidean distances among the values for the heavy metals were calculated, and then hierarchical clustering by applying Ward's method was performed on the standardized data set. In general, this form of CA is regarded as very efficient, although it tends to create small clusters. The CA results for the heavy metals are shown in Fig. 3 as a dendrogram. Fig. 3 displays three clusters for both T1 and T2. In T1, the clusters are: (1) Ni-Cr, (2) Zn-Cu, and (3) Mn-Fe; and in T2, they are: (1) Mn-Fe, (2) Cu-Cr-Zn, and (3) Ni, in total and close agreement with the PCA results for T1 and T2, respectively. However, the clusters 1 and 2 for T1, and clusters 2 and 3 for T2 join together at a relatively higher level, implying a probable common source.

3.4. Heavy metals source identification

Source identification of heavy metals is critical for pollution prevention and human health protection (Tang et al., 2013). The use of multivariate statistical techniques such as the PCA and CA, has proven to be an effective tool for extracting information on heavy metals in urban dust (Yongming et al., 2006; Tokalıoğlu and Kartal, 2006; Lu et al., 2010). In general, significant correlations between pairs of heavy metals suggest a common or combined origin, whereas weak correlations indicate different origins (Zheng et al., 2013).

Compared with the background values of world soils, the elevated concentrations of Zn, Cu, Ni, and Cr in Kermanshah atmospheric dust suggest anthropogenic sources for these elements. While Mn and Fe concentrations in most dust samples were slightly higher or even lower than their corresponding background values, indicating that they probably originated from a natural source (local soil). Also, the correlation coefficient analysis results indicate that Ni and Cr have a common source, while Mn and Fe have another common source probably. The results of the PCA and CA analysis also are consistent with these interpretations. Based on correlation coefficients, PCA and CA analyses, three main sources corresponding to groups of heavy metal pollutants, (1) Ni and Cr; (2) Zn and Cu; (3) Mn and Fe, can be identified.

A first group of elements consisting of Ni and Cr that strongly correlated in PCA and correlation coefficients analyses and were classified together in CA for T1, mainly have a common source. At the T2, however, they poorly correlated and Ni alone created a group, so that

Table 4
The rotated component matrix of the 6 heavy metals in the atmospheric dust of Kermanshah province in times T1 and T2 (PCA loadings > 0.6 are shown in bold).

Element	T1				T2			
	Component			Communalities	Component			Communalities
	1	2	3		1	2	3	
Zn	0.56	− 0.14	**0.65**	0.76	0.24	**0.88**	0.22	0.88
Cu	0.24	0.22	**0.90**	0.91	0.35	**0.71**	0.35	0.74
Ni	**0.89**	0.02	0.24	0.85	− 0.06	0.26	**0.92**	0.91
Cr	**0.88**	0.16	0.30	0.89	0.58	0.32	**0.65**	0.85
Mn	− 0.18	**0.90**	0.15	0.87	**0.89**	0.19	− 0.05	0.82
Fe	0.41	**0.81**	− 0.01	0.82	**0.87**	0.32	0.16	0.88
Eigenvalue	2.13	1.56	1.39		2.04	1.58	1.46	
Percent of variance (%)	35.50	26.08	23.27		34.07	26.27	24.34	
Cumulative percent (%)	35.50	61.58	84.85		34.07	60.34	84.68	

Extraction method: principal component analysis. Rotation method: varimax with Kaiser Normalization. Rotation converged in four and five iterations for T1 and T2.

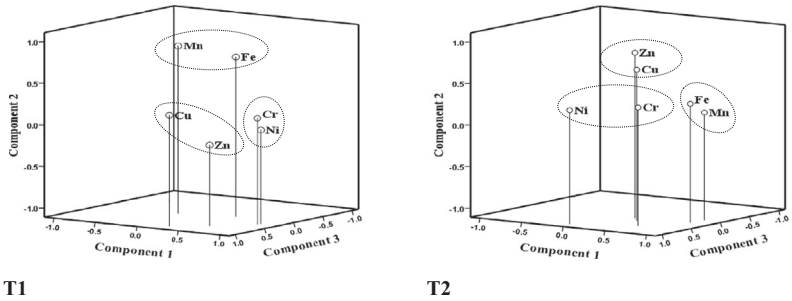

Fig. 2. PCA results in the three-dimensional space: plot of loading of the first three principal components (in times T1 and T2).

clusters 2 and 3 joined together at a relatively higher level by CA, indicating that Ni has a common source with group 2. The maximum concentrations of Ni and Cr were found in urban dust samples collected from industrial areas with heavy traffic, and the lowest concentrations were detected in samples from suburban sites with less industrial activities and traffic density. Lv et al. (2006) suggested that Ni is mainly originated from oil combustion. Ahmed and Ishiga (2006) reported that Ni is mainly of industrial sources. Wei et al. (2010a, 2010b) indicated that, main sources of Ni may include traffic, industrial, and coal combustion.

The second group of elements consists of Zn and Cu. Correlation coefficient analysis, PCA and CA analyses results for T1 indicated a strong correlation between Zn and Cu, but correlation coefficients analysis and CA in T2 indicated a strong correlation between Cu with Zn and Cr, suggesting mixed anthropogenic sources for Zn, Cu, and Cr. Moreover, PCA indicated that Zn and Cu constitute a group. The greatest Zn and Cu concentrations were found in urban dust samples from locations with heavy traffic and industrial areas, while their minimum values were observed in dust from suburban sites with less traffic density. According to previous reports (Li et al., 2001; Al-Khashman, 2007; Charlesworth et al., 2003), Zn in dust can originate from the wear and tear of vulcanized vehicle tires and corrosion of galvanized automobile parts. Copper is often used in car lubricants (Al-Khashman, 2007). It can be released to the urban environment as a result of wear of the automobile's oil pump or corrosion of metal parts which come into contact with the oil (De Miguel et al., 1997). Another possible source of Cu in the street dust is thought to be corrosion of metallic parts of cars and engine wear (Al-Khashman, 2007; Jaradat and Momani, 1999). Lu et al. (2010) showed that, Zn and Cu may have originated mainly from industry and traffic sources. According to Jiries (2001) and Al-Khashman (2004), Cr, Cu, and Zn may be from abrasion of vehicles because these heavy metals are parts of the materials for brass alloy. These metals may also be from industrial activities, confirming the previous research results (De Miguel et al., 1997; Charlesworth et al., 2003; Ahmed and Ishiga, 2006). Wei et al. (2010a, 2010b) suggested that, traffic emissions may be the major pollution sources for Zn, Cu, and Cr, while Cr may originate mainly from mixed sources of traffic instruments and industrial activities.

A third group of elements in T1 and T2, consisting of Mn and Fe strongly correlated in PCA and correlation coefficients analyses and were clearly separated from the other heavy metals in CA. This separation between Mn and Fe, and other heavy metals may suggest a mainly natural source. While, Zn, Cu, and Cr can originate from corrosion of alloys used in vehicle components, vehicle covers or other metallic surfaces and materials (Wei et al., 2010a, 2010b; Jiries, 2003), significant positive correlations and similarity of Fe with those metals could indicate that iron-containing materials and surfaces are sources of heavy metals Zn, Cu, and Cr in Kermanshah dust.

It should be mentioned that, the mineralogical association of metals in soils may be considered in their source identification studies. Based on the results of XRD of the soil samples in Kermanshah province by Heidari et al. (2008), Kermanshah soils are dominated by smectite, with different amounts of vermiculite, illite, chlorite, and kaolinite. The results of XRD and SEM (scanning electron microscopy) analyses of airborne dust samples in western Iran (including Kermanshah and Khuzestan) by Najafi et al. (2013), indicated that the mineralogy of dust samples were dominated by quartz and calcite; additionally, minor minerals including gypsum, albeit, muscovite, clinochlore, palygorskite, and dolomite. Also, they showed that the major oxides in dust storms in the study area are SiO_2 (34.82%), CaO (20.48), Al_2O_3 (8.44%), Fe_2O_3 (4.36%), and MgO (4.22%). Najafi et al. (2013) concluded that the sources of dust storm in the west of Iran are arid and semi-arid regions in Iraq, Jordan, the Arabian Peninsula, and Syria probably. These sporadic reports do not provide a significant evidence for considering the soils of the region as an important source for heavy metals presence in atmospheric dust (in particular for Zn, Cu, Ni and Cr in the current study), however, also the potential mineralogical association of the metals in dust origin soils (and rocks) could be discussed (provided that reliable information were available), prior to considering the local anthropogenic activities as the main source of atmospheric dust metals.

3.5. Pollution level assessment

3.5.1. Enrichment factor analysis

Since this is the first study on Kermanshah dust contamination, there are no historical data or background elemental concentrations reported for the area. Therefore, in this study, metal contents in world

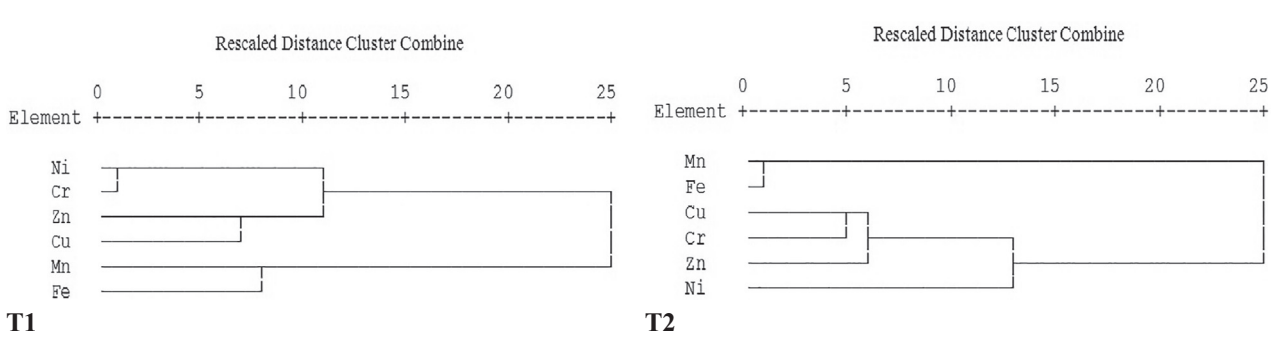

Fig. 3. Dendrogram results from Ward's method of hierarchical cluster analysis for 6 elements. Similarities have been calculated from Euclidean distance (in times T1 and T2).

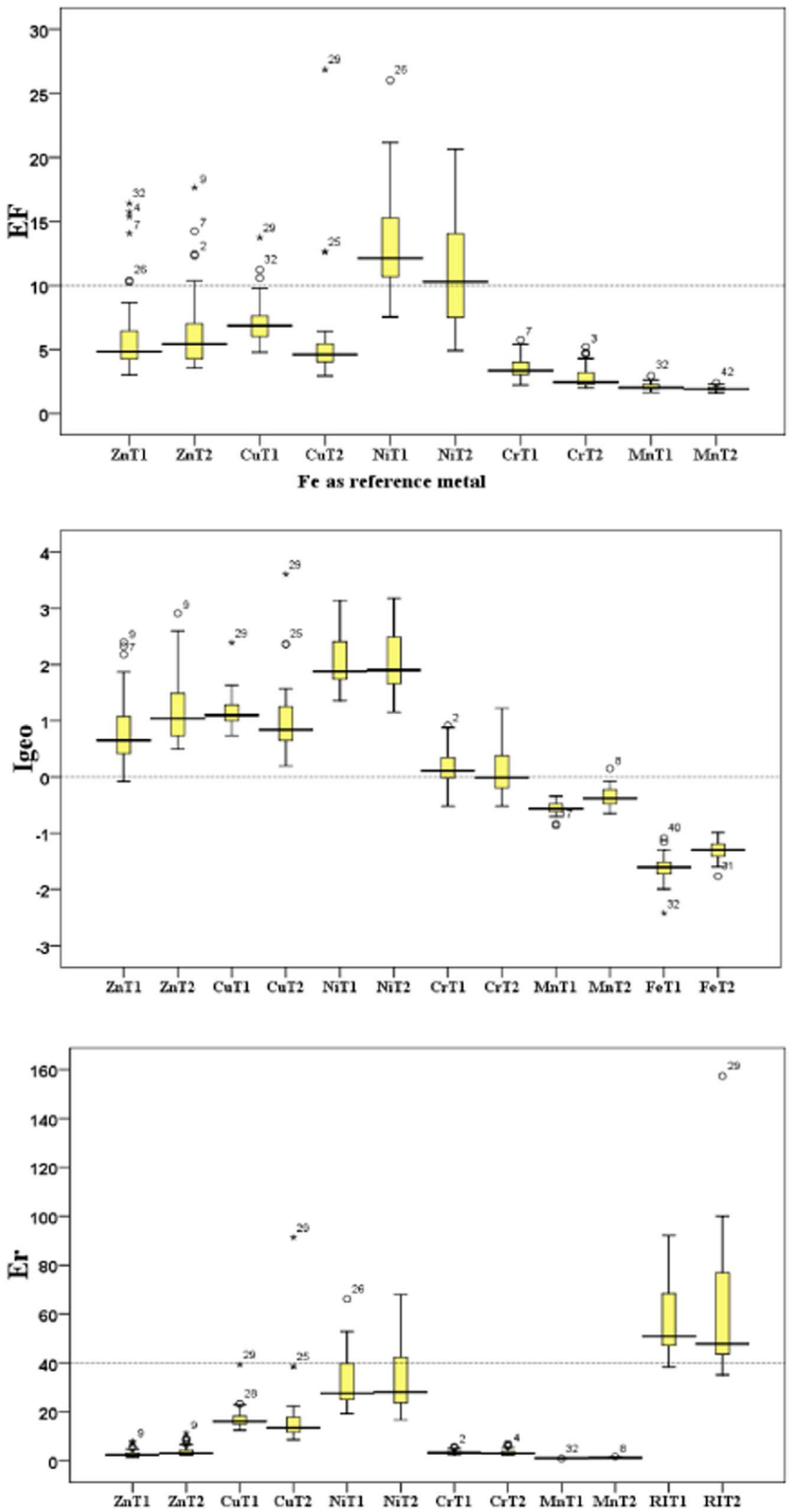

Fig. 4. Box-plots of EF, I_{geo}, E_r, and RI for heavy metals in the atmospheric dust samples of Kermanshah province in times T1 and T2.

soils were used as background values for calculating EF, as in earlier studies (Li and Zuo, 2013; Liu et al., 2014). Moreover, Fe was used as normalizer and the reference element, based on the analysis performed and references (Atiemo et al., 2011; Yongming et al., 2006; Tasdemir and Kural, 2005; Turner and Simmonds, 2006).

The EF values for heavy metals are shown in Fig. 4. The mean EFs of Zn, Cu, Ni, and Cr are close to or higher than 3, while the mean EF of Mn is less than 3. The EF value can be an indicator of natural and anthropogenic sources (Yongming et al., 2006; Lu et al., 2009). Therefore, the maximum EFs of Zn, Cu, and Ni that are higher than 10, indicate

Table 5
The values of EF, I_{geo}, E_r, and RI for studied heavy metals in the atmospheric dust of Kermanshah province in urban and suburban sites.

Sampling area		EF					I_{geo}						E_r						Ecological risk
		Zn	Cu	Ni	Cr	Mn	Zn	Cu	Ni	Cr	Mn	Fe	Zn	Cu	Ni	Cr	Mn	RI	
Urban	Minimum	3.64	3.47	5.92	2.11	1.58	0.29	0.19	1.36	− 0.52	− 0.86	− 2.42	1.84	8.57	19.22	2.09	0.82	37.90	Low
(n = 7-	Maximum	16.43	26.86	21.17	5.74	2.95	2.60	3.61	3.18	1.22	0.15	− 0.99	9.08	91.43	68.05	7	1.66	157.28	Moderate
0)	Mean	6.30	6.47	12.73	3.36	2.03	1.07	1.13	2.13	0.22	− 0.46	− 1.47	3.46	17.50	34.71	3.66	1.10	60.44	Low
	S.D.	2.91	3.12	3.96	0.94	0.25	0.60	0.45	0.48	0.42	0.18	0.25	1.72	9.92	12.42	1.15	0.14	19.88	
Suburban	Minimum	3.01	2.94	4.91	2.01	1.65	− 0.08	0.25	1.15	− 0.52	− 0.63	− 1.99	1.42	8.93	16.70	2.09	0. 97	35.14	Low
(n = 2-	Maximum	17.67	12.64	26.02	4.98	2.57	2.91	2.36	3.14	0.74	− 0.16	− 1.08	11.29	38.57	66.17	5	1.34	99.84	Low
8)	Mean	5.71	5.72	10.73	2.86	2.01	0.92	0.98	1.89	0.04	− 0.44	− 1.44	3.21	15.78	29.44	3.14	1.11	52.68	Low
	S.D.	3.48	2.38	4.33	0.66	0.22	0.67	0.49	0.46	0.25	0.14	0.22	2.08	7.04	11.41	0.57	0.11	18.37	

that these elements in atmospheric dusts mainly originate from anthropogenic sources (Liu et al., 2003). The order of EF mean values are Ni > Cu > Zn > Cr > Mn for T1, and Ni > Zn > Cu > Cr > Mn for T2. The lowest EF values were found for Mn that had 35% of EF values for T1 and 40% EF for T2 less than 2, revealing the lack of contamination with Mn as a whole and implying a significant contribution of natural sources. In case of element Cr, 92% of EF values for T1, and 98% of EF values for T2 were in the range of 2–5, with mean EF lesser than 5, representative of moderate contamination of atmospheric dusts. Zinc and Cu had 47% and 98% EFs for T1 in the range of 2–5; and 53% and 37% EFs for T2 in the range of 5–20 respectively, with mean EF higher than 5, reflecting the significant contamination of atmospheric dusts by these elements. Nickel had 94% and 6% of EF values for both sampling times (T1 and T2) in the ranges of 5–20 and 20–40, respectively, with mean EF higher than 10, indicating a significant and very high contamination. The orders of mean EFs values for urban and suburban areas are similar to the orders of EFs for T1 and T2, respectively, as presented in Table 5. Therefore, the ranges of EFs suggest anthropogenic origins for the studied metals except for Mn. Fossil fuel combustion, traffic emissions, and industrial processes are considered as the major pollution sources in atmosphere (Meza-Figueroa et al., 2007).

3.5.2. Geo-accumulation index analysis

The I_{geo} values for the heavy metals in Kermanshah atmospheric dust are presented in Fig. 4. Contamination levels were from 0 to 5, confirming the variability of atmospheric dust properties and pollution sources for the studied metals. In general, the atmospheric dusts in different areas of Kermanshah have different I_{geo} values. The order of average values of I_{geo} are similar to the order of EFs, which can also be seen as the decreasing order of their overall contamination degrees in atmospheric dusts of the study area. For Cr, the mean I_{geo} and 61% of I_{geo} values for T1 and 45% of I_{geo} values for T2 are falling into class 1 indicating unpolluted to moderately polluted status, while 39% of I_{geo} values for T1 and 53% for T2 are between 0 and 1, revealing a practically unpolluted state. The mean I_{geo} values for both sampling times (T1 and T2) obtained for Ni point to moderately to strongly polluted conditions. In this case, 65% of I_{geo} values for T1 and 59% for T2 fall into class 2 (moderately polluted), and 35% of I_{geo} values for T1 and 37% for T2 fall into class 3 (moderately to strongly polluted). The mean and distribution of I_{geo} values of elements Zn and Cu reveal the status of unpolluted to moderately polluted (71% and 31% for T1, 51% and 65% for T2) and moderately polluted (21% and 67% for T1, 36% and 29% for T2), respectively, in atmospheric dust of the study area. According to Table 5, the mean I_{geo} values are in the order of Ni > Cu > Zn > Cr > Mn for urban areas, and Ni > Zn > Cu > Cr > Mn for suburban areas, similar to the orders of I_{geo} for T1 and T2, respectively. The mean and distribution of I_{geo} values for Fe and Mn in both sampling times (T1 and T2) and in both sampling areas (urban and suburban) show that most atmospheric dust samples were practically unpolluted ($I_{geo} < 0$) with these elements. In the other word, the atmospheric dusts of Kermanshah are contaminated ($I_{geo} > 0$) by heavy metals such as Ni, Zn, Cu, and Cr (especially Ni) to some degrees, that probably originate from anthropogenic sources, whereas, Mn and Fe are not relevant as dust contaminants in the study area and are likely from natural origins.

3.5.3. Ecological risks analysis

The E_r results of the five metals (except Fe, which lacked a "toxic-response" factor) in Kermanshah atmospheric dust are shown in Fig. 4. The average E_r values of heavy metals showed decreasing order of: Ni > Cu > Cr > Zn > Mn for T1, and Ni > Cu > Zn > Cr > Mn for T2, similar to the observed orders for urban and suburban areas (Table 5), respectively. The results indicate that the mean and maximum concentrations of Ni have low and moderate potential ecological risk, respectively; while Zn, Cu, Cr, and Mn have low potential ecological risk (except for maximum concentration of Cu for T2, which have considerable potential ecological risk). Based on these results, most sampling locations show low ecological risks, which means that these metals may not have adverse effect on the ecosystem.

In general, the I_{geo} stands for "contamination" level, and the E_r for "pollution" level. Thus, it can be seen that the I_{geo} method is best applied to accumulation levels of individual metals, while the potential ecological risk index describes the ecological risk caused by pollutants (Tang et al., 2013). The results of geo-accumulation evaluation show that Ni, Zn, and Cu had moderate to strong concentrations in the atmospheric dust, but their ecological risks were relatively low. On the other hand, these metals are not at health-threatening levels, but their environmental accumulation merits more attention. To produce more comprehensive and accurate assessment results, both assessment methods should be used when evaluating the environmental quality. The calculations of the I_{geo} and the EF are both depend on the background data and the bonding forms of the metals. However, the calculations of I_{geo} are more reliable than those for EF, for the evaluation of the heavy metals contamination in dust particles, mainly for their constancy and non-significant changes as compared to EF values (Rubio et al., 2000; Ghrefat et al., 2011).

4. Conclusion

We analyzed the heavy metals status and source in atmospheric dust samples of Kermanshah province for two seasons of spring and summer, using multivariate statistical techniques combined with metal concentrations analysis and correlation analysis that has been proved to be an effective tool for source identification of heavy metals in the atmospheric dust, previously. Except for the contents of Mn and Fe, the average metal concentrations in the atmospheric dust were significantly higher than that in the background soils of the considered localities of the world. Therefore, based on the comparison of heavy metals contents of atmospheric dust in this study and background values of world soils,

the heavy metals were classified into two main groups according to their sources: natural (including Mn and Fe) and anthropogenic (including Zn, Cu, Ni and Cr). Also, based on multivariate statistical analyses, the heavy metals were classified into three main groups according to their sources.

- Zn and Cu were suggested to be from traffic activities, as well as industrial activities sources.
- Ni and Cr could be attributed to industrial activities and partly to traffic and combustion processes.
- The origins of Mn and Fe were mainly from natural sources with traces of anthropogenic influence.

To ensure more comprehensive and accurate assessment of heavy metals contamination results, three evaluation methods of enrichment factor, geo-accumulation index and ecological risk were applied.

- The calculated results of EF and I_{geo} of heavy metals revealed the identical orders of EF and I_{geo} as Ni > Cu > Zn > Cr > Mn > Fe for T1 (also for urban areas), and Ni > Zn > Cu > Cr > Mn > Fe for T2 (also for suburban areas).
- The analysis of EF also showed a moderate enrichment for Mn and Cr, and significant enrichment for Zn, Cu, and Ni (especially for Ni) in T1 and T2, and similarly for urban and suburban areas.
- The analysis of the I_{geo} results determined the levels of Mn and Fe as uncontaminated ($I_{geo} < 0$), while Zn, Cu, Ni, and Cr, were evaluated to have moderate contamination ($I_{geo} > 0$), for T1 and T2, and similarly for urban and suburban areas.
- The overall contamination levels of heavy metals were also assessed using RI. Based on RI values, heavy metals contamination were not considerable in atmospheric dust of Kermanshah.

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