

Multivariate analysis of heavy metal pollution in street dusts of Xianyang city, NW China

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Abstract Urban street dust is one of important indicators of the status of urban environmental pollution. There are many studies in capital cities, but little attention has been paid to this kind of study in medium cities, especially in China. The dust samples were collected in the district of traffic crossroads in Xianyang city, Shaanxi Province. Pb, Cd, Cu, Ni, Zn, Cr and Mn concentrations were determined by using atomic absorption spectrometry. The results indicate that street dusts in Xianyang city have elevated metal concentrations as a whole in comparison with Shaanxi Province soil, except for Mn. The content of heavy metals was comparable to those in capital cities. Correlation coefficient analysis, principal component analysis and cluster analysis were used and three main sources were identified; Mn comes mainly from soil sources; Cu, Zn and Ni are mainly derived from industrial sources combined with traffic sources; Pb, Cr and Cd have traffic sources. Heavy metal enrichment factors were calculated, which can reflect human influence degree of street dusts.

Keywords Xianyang city · Heavy metals · Street dust · Principal component analysis

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Introduction

Street dusts are the particulates that are deposited on the road, and they are a major source of pollution in the urban environment. Usually street dusts are very harmful to the human body, because the dusts usually carry toxic heavy metals (Al-Rajhi et al. 1996). The dusts that concentrate heavy metals can possibly enter the human body through the respiratory tract, which can lead to chronic poisoning because of the accumulation of heavy metals. The street dusts seriously endanger the health of urban populations, especially affecting children's physical intelligence (Harrison and Yin 2000).

Many studies about street dusts have risen in the last decades. Day et al. (1975) published the papers "Lead in urban street dust" and first proposed the terminology of urban street dust. Afterward many countries have studied heavy metals in street dusts, especially in developed economies and densely population cities (Charlesworth and Lees 1999; Li et al. 2001; Rasmussen et al. 2001). Charlesworth et al. (2003) found that, in general, the content of heavy metal in street dust is mainly affected by human activities.

There are also many researches on street dusts in China (Zhou et al. 2008; Miao et al. 2012). In China the content of heavy metal in street dust is higher than the soil background value, and space differentiation is obvious between the different functional areas (Li et al. 2008; Wei et al. 2010). Soil is the major source making up the dust that settles on city surface. Previous studies have identified the sources of heavy metals in street dusts as those associated with vehicular traffic, industrial and commercial areas (Thornton 1990) as well as weathering or building facades (Akhter and Madany 1993). So it is important to identify the source of heavy metals. However studies mainly concentrates in capital cities (Han et al. 2006), and little attention has been paid to the medium-sized cities.

The main purpose of this study is to determine seven kinds of heavy metals in the Xianyang city street dusts, and to identify their natural or anthropogenic sources by using correlation analysis, principal component analysis (PCA) and cluster analysis (CA).

Study site

Xianyang is a special place boasting a rich culture of thousands of years. It is the very place where the first Chinese Feudal Empire, Qin Empire, set its capital (221 BC). Xianyang had also served as a capital for 13 dynasties including Zhou, Han and Tang Empire. Xianyang is located in the center of Shaanxi Province and lies at latitude $34^{\circ}11'–35^{\circ}32'$ by longitude $107^{\circ}9'–109^{\circ}10'$. Xianyang has a continental season wind climate and the average annual temperature is 11.6°C , and the average yearly rainfall is 590 mm. Xianyang municipality administers two districts, ten counties and one city, covering an area of 10,196 sq km with a population of 4.83 million (XYGW 2009). The Qindu district road density reaches 0.88 km/km^2 ; the Weicheng district highway density reaches 0.7 km/km^2 . It is estimated that the whole city had 440,000 vehicles in 2011.

Methods

Sampling

Thirty street dusts sampling sites were selected in Xianyang city. The distributions of sampling sites are shown in

Fig. 1 Sampling locations in Xianyang city

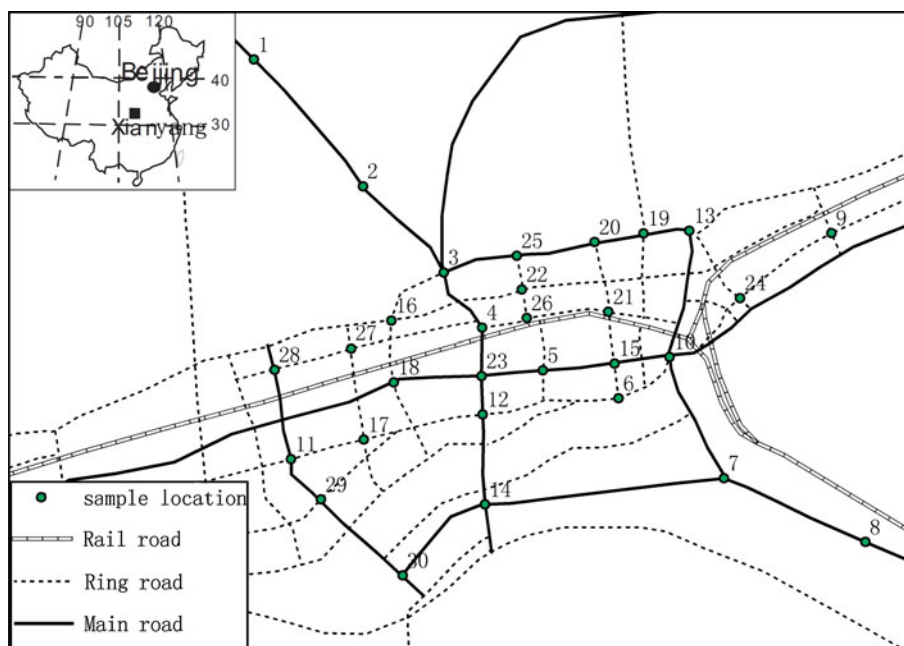


Fig. 1. Samples were collected at the street intersection by polyethylene brush and tray from five to eight points of road edges in May 2008 (Ahmed and Ishiga 2006). The sample approximately is 300 g. Dried for 2 days under the conditions of 35°C (Han et al. 2006), all samples sieved through 1 mm mesh nylon sieve to remove hair, leaves and other debris. Then the samples were grounded, homogenized and sieved through a $75\text{ }\mu\text{m}$ nylon mesh.

Analytical method

Concentrations of Zn, Ni, Pb, Cd and other trace elements have been determined by using atomic absorption spectrometry (Welz and Sperling 1999; Han et al. 2006).

Each sample (0.5 g) was put in the tetrafluoroethylene alkene crucible, adding 20 ml mixture of 10:1:10:1 HF, H_2SO_4 , HNO_3 and HClO_4 . The solution was heated on an open hot plate for about 4 h until white fumes were given off, and then the residue was redissolved in a plastic bottle with 10 ml HCl (1:1) and was diluted to 50 ml with deionized water. The atomic absorption spectroscope (novAA400) was used to determine heavy metals concentrations (Han et al. 2006). Each sample is tested three times, and the relative standard deviation is less than 5 %.

Statistical method

Principal component analysis and CA are the most common multivariate statistical methods used in environmental studies (Han et al. 2006), and play a vital role in the pollutant origin distinction (Loska and Wiechula 2003). In this study, PCA and CA were used to elucidate the latent

relationships between variables, and for investigating pollutants sources.

PCA is widely used to reduce data and to extract a small number of latent factors for analyzing relationships among the observed variables (Martin et al. 2006; Gou et al. 2007). In this study PCA with varimax normalized rotation was performed, and all principal factors extracted from the variables were retained with eigenvalues >1.0. The PC loadings were plotted to inspect the similarities observed as clusters.

The Euclidean cluster analysis technique comprises an unsupervised classification procedure that involves measuring either the distance or the similarity between objects to be clustered (Serife and Senol 2006). Hierarchical cluster analysis was used in this study. Hierarchical cluster analysis assisted in identifying relatively homogeneous groups of variables, using an algorithm that starts with each variable in a separate cluster and combines clusters until only one is left (Han et al. 2006). The linkage type used was Ward’s method and the distance method was squared Euclidean. In Ward’s method, the distance between two clusters is the ANOVA sum of squares between the two clusters added up over all the variables. At each generation, the within-cluster sum of squares is minimized over all partitions obtainable by merging two clusters from the previous generation. The sums of squares are easier to interpret when they are divided by the total sum of squares to give proportions of variance (Ward 1963).

Enrichment factor

Enrichment factor (EF) that put forward by Zoller et al. (1974) is the key parameter to evaluate the influence of

human activities on the degree of heavy metals in the sediment. It is widely used to assist the determination of the degree of metal contamination (Gordeev et al. 2004; Banat et al. 2005). The EF calculation is expressed below as:

$$EF = | Cn \text{ (sample)} / Cref \text{ (sample)} | / | Bn \text{ (baseline)} / Bref \text{ (baseline)} |$$

Cn represents the concentration of the examined element in the street dust, Bn on behalf of the examined element background value (Shaanxi Province soil background value is used here), Cref is the content of reference element in the street dust, Bref is the content of reference element in the Shaanxi Province soil. The reference element is often conservative one such as Fe, Al, Ti, Mn, Sc, etc. (Tam and Yao 1998; Reimann and Caritat 2000). Therefore, Mn was chosen as the reference element.

According to the EF, five categories of pollution (Sutherland 2000) are recognized (Table 1).

Results and discussion

Heavy metal concentrations

Background value of soil is the content of chemical elements in the natural soil without human pollution. Soil background concentrations of heavy metals are important criterion for the assessment of soil environmental quality. The descriptive statistical results of heavy metal concentrations of street dusts in Xianyang are shown in Table 2. The average values of seven kinds of heavy metals are higher than the Shaanxi Province soil background value (Chinese Environmental Protection Agency 1990). The ratio of heavy metals content mean value and background value decrease in the order of Cu > Zn > Pb > Ni > Cr > Cd > Mn. And the mean content of Cu is 5.9 times higher than background value, only the mean contents of Mn and Cd are not much higher than the background value. Besides Mn, the ranges of the content of heavy metals are also quite wide. For example, the maximum value of Cu is 17.6 times higher than the minimum value; the maximum value of Zn is 12.7 times higher than the minimum value; the maximum value of Pb is seven times higher than

Table 1 Contamination categories based on EF values

Rank	EF	The value of pollution level
1	<2	Minimal pollution
2	2–5	Moderate pollution
3	5–20	Significant pollution
4	20–40	Very high pollution
5	>40	Extremely high pollution

Table 2 Heavy metals concentrations of street dusts in Xianyang city (mg/kg)

	Pb	Cu	Cd	Zn	Ni	Cr	Mn
Maximum	164.26	1,212.59	0.23	1,029.35	188.61	253.87	733.81
Minimum	23.45	68.83	0.09	80.61	28.41	66.54	516.19
Mean value	77.30	132.17	0.13	375.38	69.54	135.63	604.36
SD	41.85	201.53	0.04	206.22	43.15	39.44	50.81
VC	0.54	1.52	0.28	0.55	0.62	0.29	0.08
Shaanxi Province soil background value	26.00	22.40	0.10	69.40	28.80	62.50	557.00

Table 3 Comparisons of heavy metals contents between different cities in street dusts (mg/kg)

City name	Population	Cd	Cu	Cr	Ni	Pb	Zn	References
London	9,227,687	6,250	61–323	ND	32–74	413–2,241	ND	Leharne et al. (1992)
Hong Kong	5,448,000	ND	92–392	ND	ND	208–755	574–2,397	Wang et al. (1998)
Hong Kong	6,733,600	ND	110	124	28.6	120	3,840	Yeung et al. (2003)
Amman	1,272,000	2.5–3.4	69–117	ND	27–32.8	219–373	ND	Jiries et al. (2001)
Shenyang	7,204,000	4.35	81.33	ND	ND	106.26	334.47	Li et al. (2008)
Xi'an	8,305,400	ND	94.98	167.28	ND	230.52	421.46	Han et al. (2006)
Xianyang	874,144	0.13	132.17	135.63	69.54	77.30	375.38	

ND no data

Table 4 Pearson's correlation matrix

	Pb	Cu	Cd	Zn	Ni	Cr	Mn
Pb		0.042 ^a	0.041	0.021	0.645	0.063	0.551
Cu	0.374 [*]		0.550	0.001	0.020	0.731	0.741
Cd	0.375 [*]	0.114		0.908	0.694	0.030	0.765
Zn	0.419 [*]	0.594 ^{**}	0.022		0.037	0.340	0.293
Ni	0.088	0.423 [*]	0.075	0.382 [*]		0.042	0.697
Cr	0.344	0.065	0.397 [*]	0.180	0.374 [*]		0.159
Mn	−0.113	−0.063	−0.057	−0.198	−0.074	0.264	

* Correlation is significant at the 0.05 level (two-tailed)

** Correlation is significant at the 0.01 level (two-tailed)

^a The left lower part is correlation coefficient; the right upper part is significance level

the minimum value. The maximum value of Mn is 1.42 times higher than the minimum value. This indicates that the Mn is possibly origin from the natural source; other elements are mainly possibly the artificial sources.

The variation coefficient (VC) of seven kinds of heavy metal content is shown in Table 2. It is stated that the VC of heavy metal content is bigger, and then the anthropogenic influence to heavy metal is bigger (Han et al. 2006). According to the VCs, heavy metals in street dusts may be divided into two categories Cu, Pb, Zn, Ni, Cd, Cr, whose VCs are greater than 0.25; Mn, whose VC is less than 0.1.

Compared with other cities (Table 3), the contents of Pb, Zn, and Cd in street dusts in Xianyang are lower than that the large city with large population, only the contents of Cu and Ni are slightly high.

Correlation analysis

To a certain extent, the correlation of heavy metals concentrations reflect the degree of similarity of these elements in causing pollution or has similar origin (Gailey and Lloyd 1985). Pearson's correlation coefficients of heavy metal elements in street dusts in Xianyang are summarized in Table 4. Mn is negatively correlated with Pb, Cu, Cd, Zn and Ni reflecting different sources of Mn. Cu, Pb, Zn, and Ni obviously showed positive correlation indicating that there may be the same source. Cd and Cr also display a positive correlation, which may suggest a common origin.

Table 5 Rotated component matrix for data of Xianyang street dust

Element	Component				Communalities
	1	2	3	4	
Zn	0.75	−0.40	0.08	0.18	0.86
Cu	0.73	−0.37	0.16	0.25	0.75
Pb	0.69	0.15	−0.43	0.37	0.78
Ni	0.62	−0.01	0.49	−0.53	0.77
Cr	0.54	0.68	0.21	−0.17	0.92
Cd	0.43	0.56	−0.50	−0.20	0.85
Mn	−0.14	0.57	0.60	0.50	0.95
Initial eigenvalues	2.46	1.42	1.11	0.82	
Percentage of variance	35.07	20.34	15.90	11.77	
Cumulative percentage	35.07	55.41	71.31	83.08	

PCA loadings >0.5 are shown in bold

Multivariate analysis

Principal components analysis

The content of heavy metals is analyzed by PCA. With varimax rotation, the factor loading values and the eigenvalues are shown in Table 5. The first and second principal component account, respectively, for 35.07 and 20.34 % of the total variance. The third and fourth principal

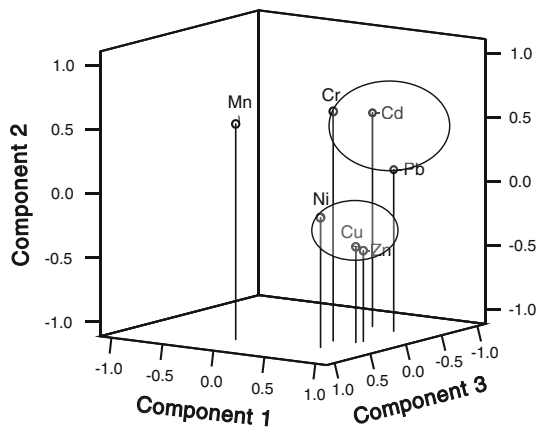


Fig. 2 PCA loading 3D plot (PC1 vs. PC2 vs. PC3) for seven heavy metals

component account, respectively, for 15.90 and 11.77 % of the total variance. So the first four principal components cumulatively account for 83.08 % of the total variance, which can reflect the majority information of the data. Factor 1 is dominated by Zn, Cu, Pb, Ni, Cr; Factor 2 is dominated by Cr, Cd, Mn; Factor 3 is dominated by Cd, Mn and Factor 4 is dominated by Ni, Mn, but the relation of Ni and Mn is a negative correlation. From Fig. 2 it can be found that the distance of Mn and other heavy metals is larger in 3D loading diagram, indicating Mn possibly has different sources from other elements. The relationship of seven kinds of heavy metals is quite obvious in the PCA 3D loading diagram (Fig. 2).

Cluster analysis

The data were standardized by means of z scores; then Euclidean distances for similarities in the variables were calculated. Finally, a hierarchical clustering by applying the Ward’s method was performed with the standardized data set. The results are shown in Fig. 3 as dendrogram. The cluster result may divide into three kinds (1) Cu, Zn and Ni; (2) Cd, Cr and Pb and (3) Mn.

Source identification

From the description of statistical analysis, the VC of Mn is the smallest, which is obviously different from other heavy metal element. Moreover, compared with the background value, the contents of Cu, Zn, Pb, Ni, Cr are larger than the background value over two times, which suggests the obvious artificial source characteristic, while the Mn content and the background value are quite close, indicating a natural source.

PCA and the CA analysis results are quite consistent, and contain much information of sources. According to the

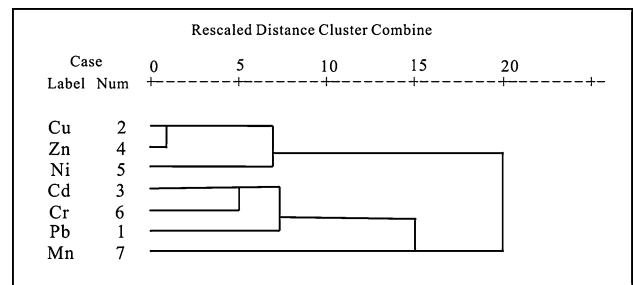


Fig. 3 Hierarchical dendrogram for seven elements obtained by War’s hierarchical clustering method (the distances reflect the degree of correlation between different elements)

PCA and the CA analysis results, Mn displays great distance with the other elements, implying that Mn has different sources with other elements. This gives additional credence to the previous analysis. Cu, Zn and Ni display a strong correlation, which is an indicator of the same sources. Pb, Cr and Cd display a strong correlation in both analyses, suggesting the same sources.

Based on the above analysis, the source of heavy metals can be divided into three categories:

1. Mn mainly comes from the natural source. In the PCA and CA analysis Mn is obviously far with other element’s distance, and its mean value close to soil background value. These both suggest that the Mn is mainly from the natural source. Mn has the obvious inverse correlation with Cu, Zn, Ni, Pb and Cd, which further supports the conclusion that it has mainly natural sources
2. Cu, Zn, and Ni mainly come from industrial source, moreover there possibly are some of the traffic sources. The average concentrations of Cu, Zn, and Ni are quite high, and have relation with heavy metal industry and chemical industry emissions (Serife and Senol 2006). For example, nearby the Xianyang pipe mill and the electrical wire factory the concentrations of Cu, Zn, and Ni are very high in No. 13 sample. Moreover Ni and Cr have the remarkable correlation, so Ni possibly also has the part originate from the alloy material used in the car body which contains Cr and Ni (Madany et al. 1994). As the widespread use of the anti-corrosion galvanized steel sheets for automobiles, a large number of zinc dust is also a source of Zn contamination (Sawyer et al. 2000)
3. Pb, Cr and Cd are mainly from the traffic source. The Pb in street dust mainly from vehicle exhausts emissions (Yeung et al. 2003). After utilization of unleaded gasoline, the contribution of vehicle exhaust to Pb particles in the atmosphere begin to decline (Zheng et al. 2004), but heavy metal has the accumulation characteristic in the soil (Kelly et al. 1996). Cr

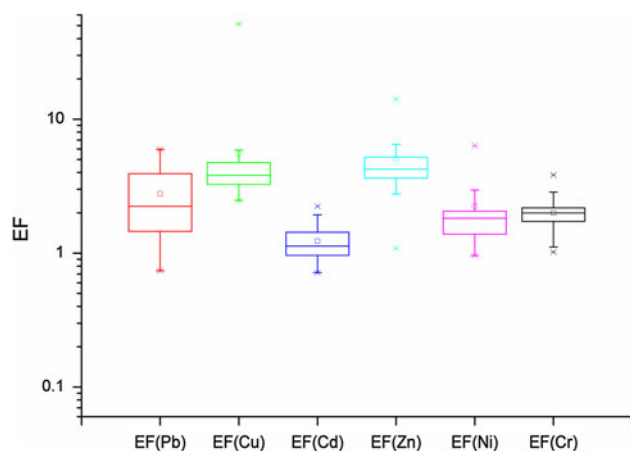


Fig. 4 Box plot of enrichment factors for metals in street dusts of Xianyang city

and Cd mainly from the aging of automobile tires wear, gasoline, and car body wear and so on. The same time, brake lining wear can also cause Cd and Pb pollution (Johansson and Westerlund 2001; Weckwerth 2001)

Enrichment factor analysis

As shown in the above analysis, Mn is one kind of conservative element in the Xianyang street dusts. The results of the other heavy metal's EF value are shown in Fig. 4. Besides Cd, other heavy metal's mean EFs are greater than 2. Moreover the maximum EFs is bigger than 3, reflecting the obvious anthropogenic pollution (Han et al. 2006). The mean EFs value in the order is: Cu > Zn > Pb > Ni > Cr > Cd, this order may also illustrate the pollution degree of heavy metal in the Xianyang street dusts. The mean EFs of Cu and Zn are bigger than 5 showing the significant pollution, the mean EFs of Pb, Ni and Cr between 2 and 5 are moderate pollution. The mean EF of Cd is smaller than 2 belonging to the minimal pollution.

Maximum value of EF may reflect the street dust pollution degree in the local area. The maximum EF of Cu is greater than 50 has achieved the extreme contamination of street dusts. The maximum EFs of Zn, Pb and Ni are greater than 5 indicating that are the significant pollution levels. The maximum EFs of Cr and Cd are also greater than 2 belonging to the moderate pollution.

Conclusion

It is feasible to judge the origin of heavy metal in Xianyang city streets through the determination of heavy metal content

using statistical analysis, correlation analysis and multi-dimensional statistical analysis. First according to the descriptive statistics and comparison with the background value, seven kinds of heavy metals were classified into two main groups according to their sources natural and anthropogenic. Then using PCA and the CA analysis, combined with the correlation coefficient analysis, the heavy metals sources were further identified. Mn is mainly derived from nature. Cu, Zn, and Ni mainly come from industrial source, while there may be some of the traffic source. Pb, Cr and Cd mainly originate from the traffic source.

The EF analysis confirms that Cu, Zn, Ni, Pb, Cr and Cd are associated with a significant anthropogenic source. According to the EF of each element, the street dusts were divided into three kinds Cu has reached an extremely high pollution degree; Zn, Pb and Ni represent significant pollution degree and Cr and Cd belong to moderate pollution.

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