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Analysis and assessment on heavy metal sources in the coastal soils developed from alluvial deposits using multivariate statistical methods

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ABSTRACT

An investigation on heavy metal sources, i.e., Cu, Zn, Ni, Pb, Cr, and Cd in the coastal soils of Shanghai, China, was conducted using multivariate statistical methods (principal component analysis, clustering analysis, and correlation analysis). All the results of the multivariate analysis showed that: (i) Cu, Ni, Pb, and Cd had anthropogenic sources (e.g., overuse of chemical fertilizers and pesticides, industrial and municipal discharges, animal wastes, sewage irrigation, etc.); (ii) Zn and Cr were associated with parent materials and therefore had natural sources (e.g., the weathering process of parent materials and subsequent pedogenesis due to the alluvial deposits). The effect of heavy metals in the soils was greatly affected by soil formation, atmospheric deposition, and human activities. These findings provided essential information on the possible sources of heavy metals, which would contribute to the monitoring and assessment process of agricultural soils in worldwide regions.

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1. Introduction

The characteristic of soil contaminated by heavy metals is commonly influenced by total heavy metal contents. Although some studies have already extended to the investigation of heavy metal fractions [1,2], it is still far from enough. These studies provided inadequate information about the bioavailability and toxicity of heavy metals [3,4]. Further studies should be focused on heavy metal sources, which will be critical to the monitoring and assessment of soil contamination [5].

There are two main sources of heavy metals in the soil: (i) natural background, which represents that the heavy metal concentration is derived from parent rocks; (ii) anthropogenic contamination, including application of agrochemicals, addition of organic amendments, animal manure, mineral fertilizer, and sewage sludge. Generally, There are more heavy metals in soils originated from anthropogenic sources than natural sources [6].

Due to urban development, the area of farmland in Shanghai has been rapidly decreasing. Meanwhile, agro-environmental pollution caused by chemical fertilizers and pesticides, industrial and municipal discharges, animal wastes, sewage irrigation, and sludge application is so serious that it would exert a potential threat on human health in Southern Shanghai [7,8]. Therefore, the identification of heavy metal sources in the present work would offer essential information on the monitoring and assessment process of agricultural soils in Shanghai.

The present study was carried out as a preliminary survey on soil contamination of the coastal plain in Shanghai. The aims of the study were: (1) to determine average regional concentrations of some heavy metals (Cu, Zn, Ni, Pb, Cr and Cd); (2) to define their natural and/or anthropogenic sources; (3) to identify their local or exotic sources causing contamination in top soils.

A multivariate statistic approach (principal component analysis, clustering analysis and correlation analysis) was adopted to assist the interpretation of geochemical data [5,9,10] and to distinguish different sources of heavy metals [11].

2. Materials and methods

2.1. Soil sampling and heavy metal determination

Twelve sampling sites were selected in southern coastal plain of Shanghai, representing an arable area of 27,838 ha. Soil parent material is alluvial formed from the Yangtze River and the East China Sea [12]. Soil samples were divided into three parts based on geo-referenced location (Fig. 1). Part 1 consisted of 8 samples (cases 1–8) collected from 4 sites located close to the city boundary. Part 2 also had 8 samples (cases 9–16) collected from 4 sites located in south of Part 1 sites. Cases 17–24 represented Part 3 collected from 4 coastal sites. Two replicates of each soil sample taken from the surface layer (0–20 cm) were analyzed, which amounted to 24 bulk cores. All the 24 soil samples were air-dried, crushed,

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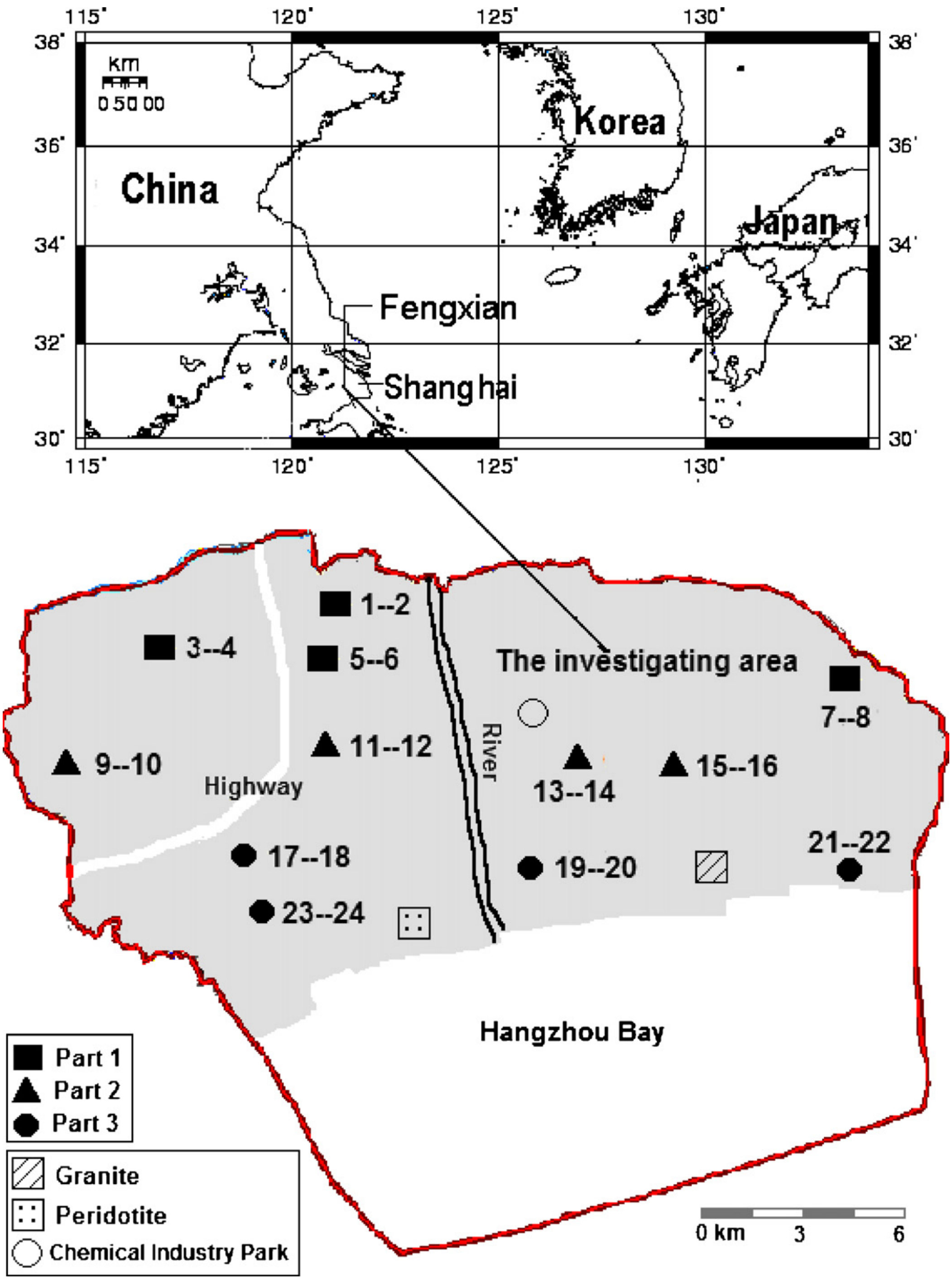

Fig. 1. Sampling sites in the agricultural area of southern Shanghai, China.

passed through a 2-mm sieve, then mixed and stored in the room temperature before analysis.

The contents of total heavy metals in soils were determined by acid digestion with a mixture of HNO_3–HF–$HClO_4$ [13], followed by flame atomic absorption spectrometry for Zn, Ni and Cr and graphite furnace atomic absorption spectrometry for Cd, Pb and Cu (Shimadzu AA6800).

2.2. Statistical analysis

The data obtained from analytical methods were treated statistically using SPSS software (version 15.0 for Windows). Principal component analysis (PCA) was employed to infer the hypothetical source of heavy metals (natural or anthropogenic). The components of the PCA were rotated by a Varimax rotation. Cluster analysis (CA) was applied to identify different geochemical groups, clustering the samples with similar heavy metal content. CA was formulated according to the Ward-algorithmic method. Results were shown in a dendrogram where procedures in the hierarchical clustering solution and values of the distances between clusters (squared Euclidean distance) are represented. Correlation matrix (CM) was used to identify the relationship between the six elements [14]. Pearson's product moment correlation coefficient was calculated in the forms of matrix [15].

Table 1
Descriptive Statistics for total heavy metal contents.

HMs[a]	Minimum ($mg\,kg^{-1}$)	Maximum ($mg\,kg^{-1}$)	Mean ($mg\,kg^{-1}$)	S.D.[b]
Cu	24.01	46.49	34.24	6.45
Zn	85.27	116.89	97.60	6.93
Ni	28.82	41.04	36.31	3.25
Pb	21.81	33.28	28.95	3.05
Cr	79.28	108.00	92.09	7.94
Cd	0.16	0.31	0.23	0.04

[a] Heavy metals.
[b] Standard deviation.

3. Results and discussion

Characteristics of the total heavy metal contents (minimum, maximum, mean and standard deviation) were presented in Table 1.

3.1. Total heavy metal contents

The mean value of the total heavy metal contents in the soils followed a descending order as: Zn > Cr > Ni > Cu > Pb > Cd (Table 1). The mean values of heavy metal contents (except that of Cd) were higher than the natural background values of Shanghai [16], but lower than the environmental quality standards for soils promulgated by National Environmental Protection Agency of China in 1995 [17].

The average Cu content with a mean of 34.24 $mg\,kg^{-1}$ was slightly higher than Soil Element Background Values of Shanghai (27.79 $mg\,kg^{-1}$), but still lower than Soil Environmental Quality Standards I (35 $mg\,kg^{-1}$). The mean of Zn content was 97.60 $mg\,kg^{-1}$,which was higher than Soil Element Background Values of Shanghai (83.81 $mg\,kg^{-1}$), but still lower than Soil Environmental Quality Standards II, proving that Zn pollution does not exit in this area. The total Ni content with a mean of 36.31 $mg\,kg^{-1}$ in all samples were higher than Soil Element Background Values of Shanghai (29.9 $mg\,kg^{-1}$), but still lower than Soil Environmental Quality Standards I (40 $mg\,kg^{-1}$). The total Pb contents with a mean of 28.95 $mg\,kg^{-1}$ in all samples were higher than Soil Element Background Values of Shanghai (21.3 $mg\,kg^{-1}$), but still lower than Soil Environmental Quality Standards I (35 $mg\,kg^{-1}$). Higher Cr content with a mean of 92.09 $mg\,kg^{-1}$ exceeds Soil Element Background Values of Shanghai (64.60 $mg\,kg^{-1}$) by 50%. The higher Cd content with a mean of 0.23 $mg\,kg^{-1}$ exceeds Soil Element Background Values of Shanghai (0.148 $mg\,kg^{-1}$) by 50%.

3.2. Principal component analysis

The results of PCA for heavy metal contents were presented in Table 2. Heavy metals were grouped into a two-component model, which accounted for 84% of all the data variation. In the rotated component matrix, the first PC (PC1, variance of 61%) included Cu, Ni, Pb and Cd, while the second PC (PC2, variance of 23%) was constituted by Zn and Cr.

PC1, including Cu, Ni, Pb and Cd, can be defined as an anthropogenic component due to their high-level presence in some soils [16].

In the sampling sites, higher content of Cu might have come from the application of Cu-contained agrochemicals [18]. Besides, industrial discharges, especially wastewater used to irrigate farmland, could be another reason for the high content [19]. Local abnormalities of total Ni content might have been resulted from sewage irrigation and stacking of municipal wastes, which might come from the river (Fig. 1) [7]. Pb content primarily came from wastewater and vehicle fume, which were possibly generated from highway (Fig. 1) [7]. The high Cd content was possibly caused by anthropogenic wastes, including sewage sludge, wastewater, and/or fertilizers and pesticides. It is worth mentioning that the accumulation of Cd in human body is primarily derived from inorganic fertilizers [20]. Our results are in agreement with previous surveys, which reported that Cd concentrations significantly increased in fertilized soils [20,21].

Another reason for the occurrence of the anthropogenic contamination of heavy metals is atmospheric deposition [8,22]. The heavy metals in the atmosphere might have come from energy production, mining, metal smelting and refining, manufacturing processes, traffic, and waste incineration, which were unfortunately widespread throughout the study area. There is a large-scale chemical industry park in the field, basically among Parts 1 and 2 (Fig. 1). Since hammered, rolled, or cast nonferrous metals (Cu, Pb, Zn, Ni, and Al) are the main production in this industrial zone, it might cause atmospheric contamination of heavy metals. Previous studies have found that atmospheric inputs of Pb, Cu and As were fairly significant to agricultural systems [23–25]. Since this study area is an alluvion along the Yangtze River, and the specific natural conditions of this area are beneficial to the atmospheric deposition [7], heavy metals deposited on the soil surface are subsequently

Table 2
Total variance explained and component matrixes for total heavy metal contents (two-components extracted).

Component	Initial eigenvalues			Extraction sums of squared loadings			Rotation sums of squared loadings		
	Total	% of variance	Cumulative (%)	Total	% of variance	Cumulative (%)	Total	% of variance	Cumulative (%)
1	3.675	61.253	61.253	3.675	61.253	61.253	3.293	54.889	54.889
2	1.367	22.780	84.033	1.367	22.780	84.033	1.749	29.145	84.033
3	0.538	8.966	92.999						
4	0.275	4.583	97.583						
5	0.092	1.538	99.120						
6	0.053	0.880	100.000						

Elements	Component matrix		Rotated component matrix	
	PC1	PC2	PC1	PC2
Cu	0.890	−0.140	0.870	0.234
Zn	0.672	0.625	0.359	0.844
Ni	0.745	−0.327	0.814	0.005
Pb	0.962	−0.119	0.927	0.283
Cr	0.371	0.868	−0.014	0.944
Cd	0.902	−0.287	0.941	0.104

Extraction method: principal component analysis; rotation method: Varimax.

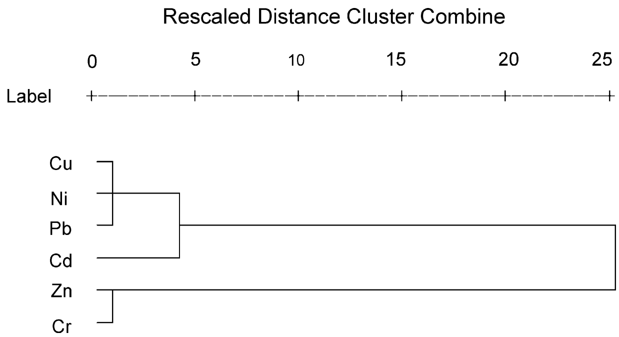

Fig. 2. Dendrogram obtained by cluster analysis for heavy metal contents.

incorporated into the soil, contributing to the overall heavy metal concentrations.

PC2 could be considered as a natural component, because the variability of the heavy metals seems to be controlled by parent rocks, moreover, Zn and Cr contents were lower than the other elements. This result suggested that the distribution of Cr and certain amounts of Zn had a lithogenic control, and these two heavy metals were included in the second principal component.

The results of probe drilling showed that the coastal part was covered with marine deposits and that stiff parent rock could be found at the depth of 300–400 m under the loose deposits [1,12]. The oldest rock in these deposits was consisted of magmatic rock [1].

Generally, the parent material exists in alluvial and alluvial–colluvial areas, and it determines Cr content, principally of calcareous nature. In natural soils, Cr is derived from the weathering of parent material and subsequent pedo-genesis [26]. In fact, the point source of contamination for Cr was not detected in the coastal plain. In the study area, Cr appears in precipitated forms in sedimentary rocks, and can possibly be related to peridotite [18].

While in this study of PCA component matrix, Zn contributed almost equally in both principal components. After being rotated, Zn showed deflection towards PC2. In the case of Zn, this element displays a combined relationship with both groups and seems to have both natural and anthropogenic sources. However, Zn in soils could have a lithogenic source as it formed a number of soluble and/or insoluble salts, according to the prevailing pedogenic processes, and in surface and subsurface horizons predominate more labile forms of Zn [27]. In addition, magmatic rock was detected in the study area, especially granite, which is the parent rock of Zn (Fig. 1).

Similarly, many authors have investigated the possible heavy metal sources in different countries. Micó [19] reported that, Cu, Cd and Pb constituted an anthropogenic component, whereas the remaining elements (Co, Cr, Fe, Mn and Ni) appeared to be associated with parent rocks. Facchinelli [11] also drew conclusions about two groups of heavy metals where Cu and Zn were associated with specific agronomic practices and Pb was derived from car exhausts, and all three kinds of metals were related to human activities. By contrast, Cr, Co and Ni were controlled by parent rocks.

In this study, it seems reasonable to conclude that Cu, Ni, Pb and Cd constitute an anthropogenic component, whereas the remaining elements (Zn and Cr) appear to be associated with parent rocks.

3.3. *Clustering analysis*

The same grouping was obtained from the clustering analysis (Fig. 2). Results revealed two clusters of elements: the first one (C1) included elements that had previously been interpreted as natural elements (Zn and Cr) while the second cluster (C2) contained the anthropogenic elements (Cu, Ni, Pb and Cd). Based on earlier discussions, the results suggested again that Cu, Ni, Pb, and Cd were mainly controlled by anthropogenic sources, while, Zn and Cr had natural sources.

Similarly, a clustering analysis calculated by sample cases was also organized and the dendrogram obtained was shown to identify the geochemical groups (Fig. 3). Cases 9, 10, 17–19 and 20 were included in Group 1; Cases 5, 8, 21–23 and 24 belonged to Group 2; Group 3 included Cases 11–16; the rest was Group 4. This group distribution corresponded to the locations of samples. Considered as a whole, Cases 5, 8–10 were local abnormal points, so they could be neglected in analysis. From this clustering analysis, Part 1 included Group 1 and 2; Part 2 included Group 3; Part 3 included Group 4, which proved that the clustered groups were in accordance with the real distribution of samples.

Total heavy metal contents were commonly high in Part 3 (Cases 17–24), especially in Cases 17–20, which mainly included Zn and Cr. Part 3 are located in coastal areas, of which saline and alkaline soils developed from alluvial deposits are predominant. Due to short land-forming time, heavy metals were deposited primarily on top soils via weathering, leaching, and/or depositing. Since the sampling sites were far from highways and chemical industry park, they were less influenced by human activities. Therefore, in this part, natural components played a dominant role.

Part 2 from middle investigated area had medium contents of heavy metals, while Part 1 from northern parts revealed high contents of heavy metals. These two parts had a longer land-forming history, and the effects of heavy metals originated from natural source were relatively smaller. The contents of Cu, Ni, Pb and

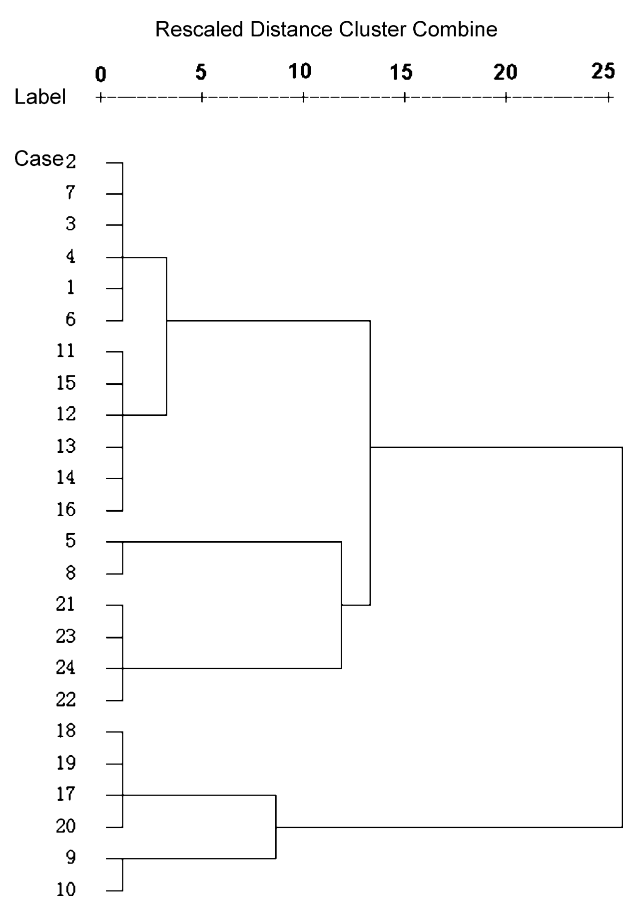

Fig. 3. Dendrogram obtained by cluster analysis for sampling sites.

Table 3
Correlation matrix of total heavy metal contents.

HMs[a]	Cu	Zn	Ni	Pb	Cr	Cd
Cu	1	0.474*	0.531**	0.826**	0.205	0.883**
Zn	0.474*	1	0.264	0.539**	0.665**	0.458*
Ni	0.531**	0.264	1	0.789**	0.070	0.621**
Pb	0.826**	0.539**	0.789**	1	0.2801	0.872**
Cr	0.205	0.665**	0.070	0.281	1	0.044
Cd	0.883**	0.458*	0.621**	0.872**	0.044	1

[a] Heavy metals.
* Significant correlation at 0.05 level.
** Extremely significant correlation at 0.01 level.

Cr were higher in these two parts, where an intensification of industrial and agricultural practices had been around for a few decades [7]. The problem of highly concentrated heavy metal contamination is possibly caused by sewage irrigation, wastewater disposal or application of fertilizer and pesticide [28]. For this group, it would be desirable to monitor these levels in order to avoid a continual increase of heavy metal contents as a consequence of human activities.

Overall, clustering analysis gave similar results, enabling the identification of the two sources of heavy metals. Therefore, multivariate analysis re-confirmed that the elements studied come from two different sources in the soils. Besides, clustering analysis by sample cases verified the fact that Part 3 (rich in Zn and Cr) was mainly affected by heavy metals originated from natural source, while Parts 1 and 2 (rich in Cu, Ni, Pb, and Cd) were influenced by human activities.

3.4. Correlation analysis

Results obtained from PCA were confirmed by the correlation analysis (Table 3). Anthropogenic metals, such as Cu, Ni, Pb and Cd, were significantly correlated. This conclusion is in agreement with other studies too [8,29].

On the other hand, natural metals (Zn and Cr) were comparatively loosely correlated as a consequence of their external sources.

The correlation coefficient matrixes between heavy metals are presented in Table 3. The total heavy metal contents except Cr were significantly correlated with each other in all the sampling sites. Total Cu content was significantly positively correlated with Ni, Pb and Cd ($P < 0.01$); Total Ni content had positive relationships with Cu, Pb and Cd ($P < 0.01$); Cu, Ni, Pb and Cd in the anthropogenic group showed more significant correlations with each other than those two elements in the natural group. By contrast, total Cr content was correlated with only Zn ($P < 0.01$), which demonstrated our group distribution above effectively. These results indicated that there existed some original relationship between heavy metals, and suggested two different possible heavy metal sources.

Besides, there is a great chance that heavy metals can bear some synergies, thus resulting in potential compound pollution [30]. These findings about relationships of total heavy metal contents could be applied to explain the behavior and the fate of heavy metals effectively and efficiently.

4. Conclusions

The results obtained in this work measured total heavy metal contents in the soils, identified heavy metal sources, and analyzed the possible causes and effects of heavy metals in the coastal soils of Shanghai, China.

The PCA performed on six heavy metals identified two principal components, which controlled their variability in agricultural soils. Cu, Ni, Pb and Cd (PC1) were related to the anthropogenic component due to their high-leveled presence in a large number of samples. PC2, including Zn and Cr, could be considered as a natural component, as the variability of these elements seemed to be controlled by parent rocks.

The same grouping was obtained from clustering analysis. Two main clusters of elements were extracted: the first one (C1) included elements that had previously been interpreted as natural elements (Zn and Cr) and the second cluster (C2) contained the anthropogenic elements Cu, Ni, Pb and Cd.

In clustering analysis on sampling sites: Cases 9, 10, 17–19 and 20 were included in Group 1; Cases 5, 8, 21–23 and 24 belonged to Group 2; Group 3 included Cases 11–16; the rest were Group 4. This group distribution corresponded to the locations of samples.

In correlation analysis, anthropogenic metals, such as Cu, Ni, Pb and Cd, were significantly correlated. While natural metals (Zn and Cr) were comparatively loosely correlated as a consequence of their exotic sources.

In this report, the involved parameter was total heavy metal content, which might not provide a complete range of information about the sources and availability of heavy metals. Therefore, extended investigation on heavy metal fractions will be developed in further studies. Information about heavy metal contents from this and other studies [18,31] can be used to assist the monitoring process of agriculture soils and the assessment system of heavy metal contamination in worldwide environment.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jhazmat.2008.08.112.

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