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The application of multivariate statistical analysis to the analysis of characteristics and assessment methods of heavy metal soil contamination in Taiwan

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Abstract : This research uses data from heavy metal soil monitoring collected in 23 countries and cities across Taiwan by the Environmental Protection Administration (EPA), Taiwan. Multivariate statistical analysis methods are applied to investigate the relationships among the variables of the eight major heavy metals, hoping to find the principal factors that affect soil contamination and the contamination tendency in Taiwan. The discussion of relationships among the eight heavy metals (i.e. chromium (Cr), Ni (nickel), Cu (copper), Zn (zinc), Hg (mercury), Cd (cadmium), As (arsenic), and Pb (lead)) can reflect the difference in the contaminations in affected areas and contamination level assessment models. The results of the factor analysis show that the principal factors affecting the contamination mechanism of the eight heavy metals can be simplified into two : (1) Highly oxidizing pollutants comprised of Cr, Ni, Cu, and Zn. The loading capacity order from high to low is Cr, Ni, Cu, and Zn, which corresponds to several rules on the periodic table. (2) The semiconductor potential pollutant factor comprised of Cd, As, and Pb. The factors, either the transition metal Cd or the typical elements, As and Pb, all have something to do with the formation of a compound semiconductor. The major mechanism for semiconductor formation is the metallic bond. As for the cluster analysis, five clusters are first divided according to the contamination characteristics. Discriminant analysis is used to classify the type of a new sample with an accuracy as high as 96.87%. This means that the clustering for the cluster analysis is acceptable. The research findings can serve as reference material for relevant entities in the planning and application of soil remediation strategies.

Keywords : Soil metal, multivariate statistical analysis, factor analysis, discriminant analysis, soil pollution.

Introduction

In the last thirty years or so, with the ever growing size and diversity of industrial activities in Taiwan, the resulting vast volume of waste and pollutants generated, soil contamination of organic pollutants and heavy metals in particular, has become a widely recognized environmental issue. Soil plays a fundamental role in Taiwan's agricultural economy and provides a source for people's livelihood. Soil can absorb, exchange, oxidize, and retain matter and therefore, it can help reduce the impact of pollutants on the environment as it degrades the waste generated during a natural cycle. However, when the build-up of pollutants is beyond the soil's self-purification capacity, the result is soil contamination. Agricultural production will be impacted and people's health and lifespan will be threatened. To tackle the aggravating soil contamination problem and conserve the valuable soil resource, countries worldwide have started to engage in soil investigation, contamination prevention, and remedial actions.

Multivariate monitoring methods that consider all available data simultaneously can extract key information about the relationships and combined effects of air pollutants. When failures occur in air quality management systems, univariate monitoring methods are often inadequate in identifying causes because the signal-to-noise ratio is very low in each air pollutant measurement. But multivariate monitoring can improve the signal-to-noise ratio through averaging, resulting in a more realistic evaluation of the environmental context[1–4]. In the field of chemometrics, multivariate statistical techniques have become one of the most active research tools in modeling and analysis over the last decade[5,6]. However, to the authors' knowledge, only limited research on the effectiveness of multivariate models for the assessment and management of air pollution has been conducted thus far[7,8].

The multivariate statistical techniques such as cluster analysis (CA), factor analysis (FA), principal component analysis (PCA) and discriminant analysis (DA) have widely been used as unbiased methods in analysis of water quality data for drawing meaningful information[9–12]. The multivariate treatment of data is widely used to characterize and evaluate surface and freshwater quality and it is useful for evidencing temporal and spatial variations caused by natural and anthropogenic factors linked to seasonality[13].

This research uses data from heavy metal soil monitoring collected in 23 counties and cities in Taiwan and applies multivariate statistical analysis methods to investigate the relationships among the variables of the eight major heavy metals (i.e. Cr, Ni, Cu, Zn, Hg, Cd, As and Pb). The goal is to find the principal factors that affect soil contamination and the tendency of contamination in Taiwan. As the discussion of relationships among the eight major heavy metals can reflect the difference in contamination in affected areas and in contamination level assessment models, the research hopes to help establish an assessment method that best reflects the soil's heavy metal contamination in Taiwan and can be used for grading. The research starts with an analysis of the characteristics of soil contamination by the eight heavy metals in 23 counties and cities in Taiwan; the findings are expected to serve as reference for relevant soil contamination prevention and for the management authorities in Taiwan in developing remediation technologies and administrative management.

Methodology :

Selection and source of soil's heavy metal data :

The heavy metal data used in this research is sourced from the EPA's environmental data warehouse system website in Taiwan (http://edw.epa.gov.tw/topicSoil.aspx). The data used in this research was collected during a time period from January 2013 to December 2015. Any incomplete or missing data regarding the eight major heavy metals are excluded; hence, the number of data entries for each of the counties and cities differs. This research uses a total 6176 data entries, all completed with variable information on the soil's heavy metals. Fig. 1 shows the location of the 23 counties and cities in Taiwan.

Statistical analysis – factor analysis :

In order to select the elements to be included in the FA, a minimum of 70% of the samples need to have measurable levels of an element. In principle, FA actually groups the elements whose concentrations fluctuate together from one sample to another and separates these elements into factors[14–17]. Factor analysis is used for source apportionment in environmental data with the argument that elements that fluctuate together have some common characteristics. Ideally, each extracted factor represents a source affecting the samples. The factor analysis was conducted with the Statgraphics Plus program package (Statgraphics Manual 3.1 1997). The initial components were rotated using the varimax method to obtain final eigenvectors with the most representatives of individual sources of variation. Although there are no well-defined rules on the number of factors to be retained, usually either factors that are meaningful or factors with eigenvalues greater than one are retained. In theory, irrelevant factors have zero eigenvalues and eigenvalues less than one indicate that a factor contributes less than a single variable. The physical meaning of the factors must be interpreted by observing which elements or variables display a high (≥0.25) loading within the factor. Loadings of less than 0.25 in absolute value may be dominated by random errors. There is not a set rule for the selection of the number of factors, but in application, the selected number of the factors must explain at least 70% of the total variance. Then the data are screened for outliers using their factor scores. The magnitude of a factor's (i.e. source's) influence on a specific sample is given by the factor score for that sample[18,19]. The factor score is the number of standard deviations from the mean of that factor as averaged over all the samples; in other words, it is the value of the factor. An average contribution from the factor results in a score of zero, a larger than average contribution results in a positive score and a lower than

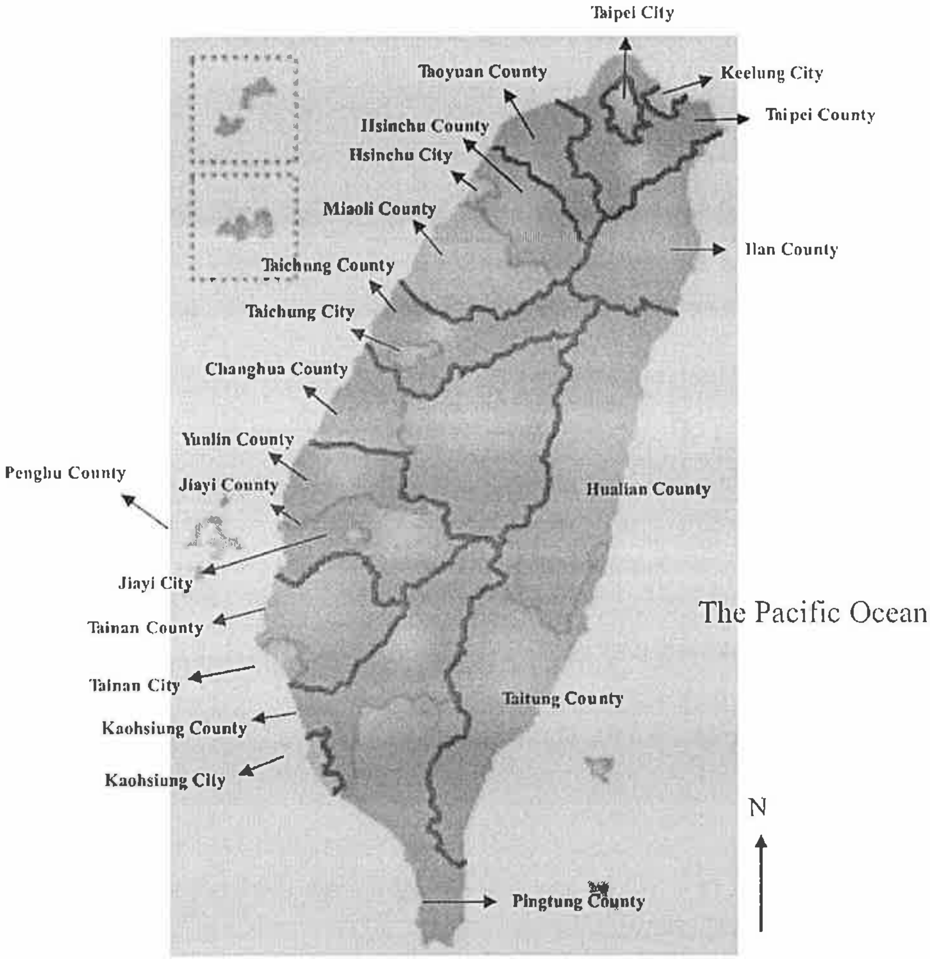

Fig. 1. Map showing the locations of the counties and cities in Taiwan.

average contribution results in a negative score. Factor scores greater than one indicate a strong influence of that source or factor on that individual sample.

Cluster analysis :

Cluster analysis is an exploratory data analysis tool for solving classification problems. Its objective is to sort cases into groups, or clusters, so that the degree of association is strong between members of the same cluster and weak between members of different clusters. Each cluster thus describes, in terms of the data collected, the class to which its members belong; and this description may be abstracted through use from the particular to the general class or type. Hierarchical agglomerative clustering is the most common approach as it provides intuitive similarity relationships between any one sample and the entire dataset. It is typically illustrated by a dendrogram (tree diagram)[20]. The dendrogram provides a visual summary of the clustering processes, presenting a picture of the groups and their proximity, with a dramatic reduction in dimensionality of the original data. Additionally, cluster analysis helps in grouping objects (cases) into classes (clusters) on the basis of similarities within a class and dissimilarities between different classes. The class characteristics are not known in advance but may be determined from the analysis. The results of CA help in interpreting the data and indicate patterns[21,22].

Discriminant analysis :

Discriminant analysis is used to determine the variables that discriminate between two or more naturally occurring groups. It uses raw data to construct a discriminant function for each group (Wunderlin *et al.*) as in eq. (1):

$$f(G_i) = k_i + \sum_{j=1}^{n} w_{ij} p_{ij} \quad (1)$$

where i is the number of groups (G), k_i is the constant inherent to each group, n is the number of parameters used to classify a set of data into a given group, w_j is the weight coefficient assigned by DA to a given selected parameter (p_j). In this case study, three groups of temporal (three seasons) and spatial (three sampling regions) evaluations have been selected and the number of analytical parameters used to assign a measure from a monitoring site into a group (season or spatial) has been taken as n. Discriminant analysis is applied to the raw data by using the standard, forward stepwise and backward stepwise modes to construct discriminant functions to evaluate both the spatial and temporal variations in air quality. The temporal (season) and the spatial (site) were the grouping (dependent) variables, while all the measured parameters constituted the independent variables.

Results and discussion

Selection of the factor analysis results :

In computing the factor analysis, this research uses the varimax for orthogonal rotation to explain the characteristics of the factor number. Table 1 shows the results : two factors have eigenvalues greater than one, and the explained, accumulative variance of the two common factors is 59.634%. The eigenvalues of the common factors are 3.470 and 1.301. As shown in Table 2, the KMO (Kaiser-Meyer-Olkin measure of sampling adequacy) is 0.806, which is greater than 0.5; according to Kaiser, factor analysis is suitable in this case.

Table 1. Results of factor analysis and the variance explained for the eight heavy metal variances

Components	Initial eigenvalues	% of total variance	Cumulative variance (%)
1	3.470	43.371	43.371
2	1.301	16.263	59.634
3	0.966	12.077	71.711
4	0.758	9.479	81.190
5	0.671	8.386	89.576
6	0.357	4.460	94.036
7	0.295	3.692	97.728
8	0.182	2.272	100.000

Table 2. KMO and Bartlett's test table

Kaiser-Meyer-Olkin measure of sampling adequacy		0.806
Bartlett test	Chi-square test	19490.909
	Degree of freedom	28
	Significance	0.000

Selection of factors :

As mentioned in the previous section, the number of eigenvalues greater than one can determine the number of principal factors. The component matrix of the number of principal factors after the orthogonal rotation can be used to select the variables between the factors. Table 3 shows the component matrix after the orthogonal rotation. The matrix after rotation explains the characteristics of the two factors, which can be used to describe the major causes affecting the quality of heavy metals in the soil and to discuss the characteristics and differences of the factors.

Table 3. Factor loading matrix for the eight heavy metals

Components	Factors 1	Factors 2
Cr (chromium)	0.871	0.077
Ni (nickel)	0.864	0.205
Cu (copper)	0.845	0.014
Zn (zinc)	0.813	0.308
Hg (mercury)	0.261	−0.080
Cd (cadmium)	0.193	0.747
As (arsenic)	−0.269	0.736
Pb (lead)	0.432	0.525

Explanation of the factors :

Table 3 shows that there are two principal factors affecting the quality and characteristics of the soil's heavy metals. The three-dimensional distribution diagram (shown in Fig. 2) of the major factors that affect the quality of the soil's heavy metals indicates that there are two axes; Cr, Ni, Cu and Zn fall on one axis and are classified under Factor 1; Cd, As, and Pb are categorized as Factor 2 and they fall on the other axis. Despite falling on an axis of its own, mercury has an unapparent eigenvalue (lower than 1) so that it is not explained in this research. The following provides complete information on the characteristics of the two factors.

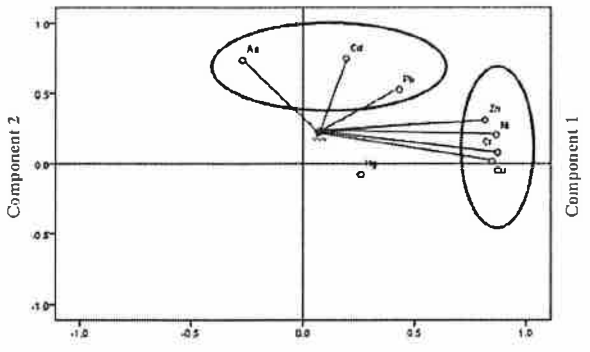

Fig. 2. Three-dimensional distribution of factor analyse for soil 8 heavy metals.

(i) Factor 1 :

As shown in Table 3, Factor 1 is comprised of four variables including Cr, Ni, Cu, and Zn; their total variance can reach 43.371%, as shown in Table 1. Its impact can take up almost half of the explanation for the soil's cumulative variance. The factor represents the four heavy metals commonly found as contaminants in soil. When they are in the oxidation state, they are highly oxidized, can affect soil quality, and furthermore, threaten the surrounding environmental agents, such as the ground surface, underground water, air, or even the human body. As a result, what makes up the factor can be referred to as a "highly oxidizing pollution factor".

(ii) Factor 2 :

Table 3 shows that Factor 2 is made up of three variables : cadmium, arsenic, and lead; Table 1 shows its total variance is 16.273%. The three heavy metals are either transition metal Cd or the typical elements. As and Pb, and have something to do with the formation of the compound semiconductor. The key mechanism of the semiconductor formation is the metallic bond. The formation of the compounds of these heavy metals can seriously contaminate and impact the environment and agents. Therefore, it can be referred to as the "semiconductor potential pollution factor".

Analysis of the properties of the eight major heavy metal soil pollutants–cluster analysis :

A two-phase clustering process is used. First, hierarchical clustering is used for general clustering; then, the K-mean method is used to test the cluster numbers. Finally, five clusters are used to categorize the characteristics of the eight heavy metal contaminants. Fig. 3 shows the relationship between the clusters and the factors. Moreover, Table 4 provides a glance at the representativeness and percentage of the clusters in the 23 counties and cities. It also shows the cluster properties of the heavy metals in the counties and cities.

(1) Cluster 1 :

As shown in Fig. 3, Factor 1's (highly oxidizing pollution factor) score for Cluster 1 is the fourth highest among the clusters while Factor 2's (semiconductor potential pollution factor) score is the third highest; that is, the distribution concentration of this cluster of metals is not high. In general, the concentration levels of the metals in this cluster indicate low levels of contamination, which can be proven by the factors' scores. Although this cluster does not have as high levels of contamination as the others, under soil contamination regulations, there is still a large volume of soil considered to have been contaminated. Generally speaking, the soil samples in this cluster mainly come from central Taiwan; then, from northern Taiwan. Except for Kaohsiung County, comparatively fewer soil samples in this cluster are from southern Taiwan. Since the concentration level of the cluster is lower in Factor 1, this cluster can be referred to as the "low-concentration, highly oxidized cluster".

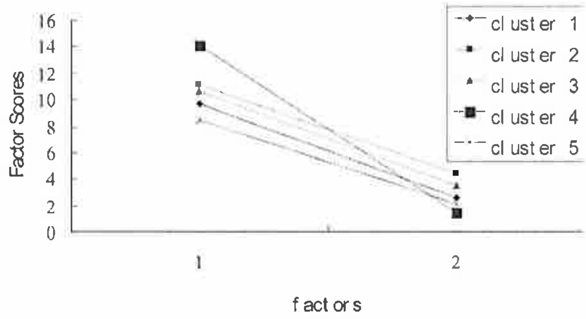

Fig. 3. Relations between the clusters and factors.

(2) Cluster 2 :

As shown in Fig. 3, Factor 1's (highly oxidizing pollution factor) score for this cluster is the second highest, while Factor 2's (semiconductor potential pollution factor) score is the highest. This means that the distribution concentration of the cluster is generally higher. Whether judged from Factor 1 or Factor 2 scores, the contamination levels of metals in this cluster are generally high. The

Table 4. Representativeness and percentage of the clusters in the 23 counties and cities

Clusters County or City(%)	Cluster 1	Cluster 2	Cluster 3	Cluster 4	Cluster 5
Taipei County	30.7%	4.5%	6.3%	1.1%	57.4%
Taipei City	0%	0%	0%	0%	100%
Keelung City	2.7%	0%	0%	0%	97.3%
Yilan County	0%	0%	0%	0%	100%
Taoyuan County	23.4%	3.8%	2.8%	0%	68.0%
Hsinchu County	0%	0%	0%	0%	100%
Hsinchu City	48.1%	20.9%	0.7%	5.0%	25.3%
Miaoli County	16.2%	0%	2.3%	0%	81.5%
Taichung County	46.6%	8.1%	13.0%	0%	29.3%
Taichung City	18.9%	0.7%	0%	0%	80.4%
Nantou County	0%	0%	0%	0%	100%
Changhua County	66.8%	10.4%	5.7%	1.9%	15.2%
Yunlin County	0%	0%	0%	0%	100%
Jiayi County	0%	0%	0%	0%	100%
Jiayi City	0%	1.7%	0%	0%	98.3%
Tainan County	10.3%	1.1%	7.2%	0.3%	81.1%
Tainan City	16.7%	1.5%	12.1%	0%	69.7%
Kaohsiung County	26.8%	3.3%	8.7%	0%	61.2%
Kaohsiung City	1.8%	0%	0%	0%	98.2%
Pingtung County	7.6%	0%	1.5%	0%	90.9%
Hualian County	0%	0%	0%	0%	100%
Taitung County	0%	0%	0%	0%	100%
Penghu County	0%	0%	0%	0%	100%
Number of the soil data entries	1522	372	186	71	4025
Characteristics of each cluster	Based on the data of Changhua County	Based on the data of the second factor pollution	Higher levels of the arsenic concentration in Central Taiwan Science Park (Taichung County)	Based on the data of the first factor (Hsinchu City)	Based on the data of the administrative zones with lower contamination levels

soil samples in this cluster are from central and northern Taiwan; however, the percentage of this kind of soil is not high within the central and northern regions. Offshore and eastern areas do not have the cluster. It can be concluded that the contamination of soil in this cluster is caused by local industrial activities. This cluster can be referred to as the "highly contaminated cluster".

(3) Cluster 3 :

As shown in Fig. 3, the factor (highly oxidizing pollution factor) score of this cluster is the third highest while Factor 2's (semiconductor potential pollution factor) score is the second highest. This means that the distribution concentration of this cluster is relatively higher. Whether judged from Factor 1 or Factor 2's scores, the contamination level or potential contamination of Cluster 3 is higher than that of Cluster 1 and lower than that of Cluster 2. The soil samples in this cluster are from a number of administrative regions across northern, central, and southern Taiwan, with the majority from the central area, where air pollution in Central Taiwan Science Park is to blame for the soil contamination. The seven soil heavy metal concentration levels of the two factors in this cluster are lower than those in Cluster 2. This cluster can be referred to as the "intermediately contaminated cluster".

(4) Cluster 4 :

As shown in Fig. 3, Factor 1's (highly oxidizing pol-

Table 5. Results of Discriminant Analysis

Discriminated cluster/ Actual cluster	Cluster 1	Cluster 2	Cluster 3	Cluster 4	Cluster 5	Percentage of accurate discrimination (%)
Cluster 1	1463	31	26	0	2	(1463/1522)×100 = 96.12
Cluster 2	5	366	1	0	0	(366/372)×100 = 98.39
Cluster 3	2	1	183	0	0	(183/186)×100 = 98.39
Cluster 4	0	7	1	63	0	(63/71)×100 = 88.73
Cluster 5	59	0	0	0	3966	(3966/4025)×100 = 98.53
Total	1529	405	211	63	3968	(6041/6176)×100 = 97.81

lution factor) score of this cluster is the highest, while Factor 2's (semiconductor potential pollution factor) score is the lowest. This means that there is a difference between the factors. A comparison of Factor 1's contamination percentage in Clusters 1, 2 and 4 shows that although Cluster 4 has the highest levels of Factor 1 contamination (with the highest Factor 1 score), the range of contamination and sites affected are not as obvious as those in Clusters 1 and 2. As for administrative regions with lower levels of contamination, such as the offshore islands, Yilan County, and eastern Taiwan, even though the contamination level of Factor 2 is not apparent, no representative samples can be collected because the contamination characteristics do not fit those of Factor 1. The cluster can be referred to as the "high-concentration, highly oxidized cluster".

(5) Cluster 5 :

As shown in Fig. 3, Factor 1's (highly oxidizing pollution factor) score of this cluster is the lowest, while Factor 2's (semiconductor potential pollution factor) score is the second lowest (or the fourth highest). This means that the distribution concentration of this cluster is generally lower. The soil is commonly distributed across Taiwan's administrative districts, especially in eastern and north-eastern areas and Penghu County, where the population is lower and the potential of industrial pollution is lower. These areas are therefore less prone to extensive contamination. This cluster can be referred to as the "low-concentration contaminated cluster".

Discriminant analysis :

Discriminant analysis is a method used to objectively determine the type of a new sample based on the properties observed and figures collected, given a classification system. Table 5 shows the discriminant analysis results of the five clusters.

Table 5 shows the percentage of accurate discrimination between the discriminated cluster generated from the discriminant function and the actual cluster generated from the cluster analysis. The percentages of accurate discrimination of the five clusters are rather high. Cluster 1 reaches 96.12%; Cluster 2 reaches 98.39%; Cluster 3 is 98.39%; Cluster 4 is 88.73% and Cluster 5 is 98.53%. This means that all five clusters can provide accurate discrimination. Clusters 2, 3, and 4 have higher percentages of accurate discrimination. As mentioned previously, Cluster 2 is highly contaminated cluster with high Factor 1 and Factor 2 scores; Cluster 3 is intermediately contaminated cluster; and Cluster 4 is "high-concentration, highly oxidized cluster" with a high Factor 1 score. The soil samples are from places with more serious soil contamination with heavy metals, and the percentages of accurate discrimination are all higher than 98%. This means that the likelihood of inaccurate discrimination for higher levels of contamination is low; samples from areas with high levels of heavy metal contamination where soil is affected much more provide more traces of the contamination characteristics so that the discrimination accuracy is higher. Hence, the clustering for the five-cluster analysis is acceptable.

Conclusion

(1) This study uses soil heavy metal data from 23 counties and cities across Taiwan as well as multivariate statistical methods to examine to investigate the relationships among the variables of the eight major heavy metals. Contamination characteristics can be simplified into two groups : (i) being a highly oxidizing pollution factor comprised of chromium, nickel, copper, and zinc; and (ii) having a semiconductor potential pollution factor, comprised of Cd, As, and Pb.

(2) The four heavy metals in Factor 1 happen to fall into the fourth series in the periodic table; they are the

transition metals in the first transition series. These metals are characterized as strong oxidants. The loading capacity order from high to low is Cr, Ni, Cu, and Zn. Their atomic radiuses on the periodic table also show a progressive decline.

(3) The three heavy metals in Factor 2, the transition metal cadmium and the typical elements, arsenic and lead, have something to do with the formation of the compound semiconductor and the major mechanism for semiconductor formation is the metallic bond.

(4) Concerning the cluster analysis, the contaminated soil in Taiwan can be classified into five clusters based on the characteristics of the contamination : the "low-concentration, highly oxidized cluster", the "highly contaminated cluster", the "intermediately contaminated cluster", the "high-concentration, highly oxidized cluster" and the "low-concentration contaminated cluster".

(5) This research uses the discriminant analysis to determine the percentages of accurate discrimination from the cluster analysis results. The percentages of accurate discrimination level of the five clusters can reach 97.81%; this means that these five clusters can be used to accurately represent the clusters.

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