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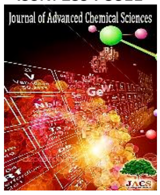

Source Identification of Heavy Metal Contamination in Ground Water

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ABSTRACT

A comprehensive groundwater heavy metal contamination in groundwater study was conducted in Virudhunagar district, Tamilnadu aiming to investigate the impacts of urbanization on the aqueous distributions of these elements. Groundwater samples were collected from 8 sampling sites and analyzed for heavy metals such as Zn, Cu, Pb, Cd and Fe. The results were compared with BIS and WHO standards. The concentrations of heavy metals were within the desirable limit at all the sampling sites. The multivariate statistical analysis revealed that Fe showed almost independent pattern among the other studied metals. Variation of Zn, Cu, Pb, and Cd concentrations is controlled by anthropogenic intense agriculture activities. The concentration of Fe is mainly affected by natural parent material and human activities. This study keep scores the value of multivariate statistical analysis for evaluation and interpretation of the data with a view to stimulate better policy outcomes and decision-making that positively impact water quality and thus prospectively diminishes the pollution caused by hazardous toxic elements in environment.

1. Introduction

The irrepressible human curiosity and the unquenchable thirst for knowledge are the fundamental basis for scientific development. A major part of innovations in scientific and technological development has been directed towards the elevation of human comforts, thereby increasing the standard of living in the society. This led to increase in industrialization. Some of the important improvements in our standard of living that can be attributed to the application of science and technology include production of more and better quality food, elimination of various infectious diseases, invention of new and faster communication systems, creation of reliable and faster transportation, supply of safe water, invention of machines to replace human and animal power, minimizing water-borne diseases through improved water technology and mitigation of bad effects due to natural disasters such as droughts, floods and volcanic eruptions. Consequent to these improvements, disturbing side effects such as environmental pollution, deforestation, urbanization and evolution of new organisms resistant to control have emerged. These effects are considered as potential threats to environment and to human beings.

The ground water is defined as water that is found underground in cracks and spaces in soil, sand and rocks. In the hydrological cycle, less than 0.1% of the metals are actually dissolved in the water and more than 99.9% are stored in sediments and soils [1,2]. The rate of depletion of ground water levels and deterioration of ground water quality is of immediate concern in major cities and towns of the country [3-20]. Heavy metals are among the major pollutants of water sources. Metals in natural waters are induced from various sources. Natural geological weathering of rocks and soils, directly exposed to surface waters, is usually the largest natural source. Several studies have shown that metals exist at low concentrations in natural waters, partially in soluble ionic forms and partially forms bound to inorganic or organic particulate matters and their toxicity can be attributed mainly to their soluble forms .Besides the natural processes, metals may enter into the aquatic system due to anthropogenic factors such as mining operations, disposal of industrial wastes and applications of biocides for pest. Anthropogenic sources of elemental contamination and pollution released into the environment have been summarized by many researchers.

Heavy metals are some of the main source of toxicity problems in the aquatic environment when they occur above the threshold concentrations. The behavior of heavy metals in the environment depends on their inherent chemical properties. Trace metal contaminations are important due to their potential toxicity for the environment and human beings. Some of the metals are the essentials for metabolic activity in organisms to sustaining aquatic biodiversity, but there is a narrow gap between their essentiality and toxicity. Toxic heavy metals can accumulate in the bodies of aquatic organisms, including fish, making them unfit for human consumption. The most toxic heavy metals are such as chromium, nickel, lead, cadmium, and arsenic. Cr (VI), nickel and cadmium are carcinogenic; arsenic and cadmium are teratogen and the health effects of lead include neurological impairment and malfunctioning of the central nervous system [21]. Although some heavy metals such as iron, manganese, cobalt, copper and zinc are essential micronutrients for fauna and flora, they are dangerous at high levels [21-22].

Furthermore, multivariate methods have been used to compare the results coming from the principal component analysis carried out on the concentration data with the experimental indicator variogram applied to some categorical information, in order to relate the concentration of heavy metals to the geology and land use of the area [23].

The main objective of this study are, 1. Assessing the water contamination by heavy metals and prioritizing contaminated areas for further investigation and 2. Distinguishing between natural and anthropogenic sources of the studied metals by multivariate statistical methods.

2. Experimental Methods

2.1 Study Area

Virudhunagar district is located in Tamilnadu, India. It lies between Latitude 9.012582° N to 9.045470° N and Longitude 77.024953° E to 7.8018971° E. The district is bounded by Sivagangai District and Madurai District on the north, Tirunelveli District and Tuticorin District to the south and Ramanathapuram District on east and Kerala state to the west and Theni District to the North West. The location of the study area is shown in Fig. 1. Total area of Virudhunagar district is 4243.23 sq km and the

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district is divided into 8 Taluks. The eight taluks are Aruppukkottai, Virudhunagar, Rajapalayam, Sattur, Sivakasi, Srivilliputtur, Kariapatti and Tiruchuli. The district receives the rainfall under the influence of both southwest and northeast monsoons. The Northeast monsoon chiefly contributes to the rainfall in the district. The mean annual rainfall of study area is about 800 mm. The relative humidity is on an average between 65 and 85%.

Virudhunagar district is characterized by relatively high level of groundwater development in both hard rock and sedimentary aquifers. Occurrence, movement and storage of groundwater are influenced by lithology, thickness and structure of the rock formation. The presence of black clayey soils has resulted in reduced natural recharge to groundwater system. It has also resulted in water quality problem. Virudhunagar district is leading in the country in the match industry, fireworks and printing, mostly concentrated in and around Sivakasi. Virudhunagar is the main market for oil chicory, coffee seeds, dry chillies and pulses. There are two trade centers (Godowns) in the District one in Virudhunagar and another in Rajapalayam. Ginning factories, spinning mills, power loom and hand loom industries are also present in the district at Rajapalayam the second big weaving town in the state after Coimbatore. Map of the study area is given in Fig. 1. The description of sampling sites is given in Table 1.

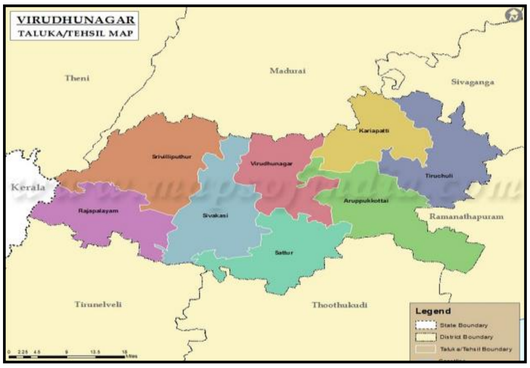

Fig. 1 Map of the study area

Table 1 Description of eight sampling sites in Virudhunagar District

Site No	Sampling Sites
S_1	Aruppukkottai
S_2	Kariapatti
S_3	Rajapalayam
S_4	Sattur
S_5	Sivakasi
S_6	Srivilliputtur
S_7	Tiruchuli
S_8	Virudhunagar

2.2 Collection and Analysis of the Samples

The groundwater samples are collected from nine different sampling sites in previously acid-washed 5 litre high density polyethylene (HDPE) containers and filtered through pre-washed 0.45 μ millipore nitrocellulose filters on the sampling day. The initial portion of the filtration was discarded to clean the membrane, and the following ones were acidified to pH< 2 with ultra-purified 6 M HNO_3 and then stored in pre-cleaned HDPE bottles for trace metal analyses. Cleaning of plastic bottles was carried out by soaking in 15% (v/v) HNO_3 for 24 hrs. and then rinsing with deionized water (analytical grade). Acid-cleaned polyethylene gloves were used while handling all plastic and glass ware and analytical reagents were used throughout the determinant processes. The samples are then refrigerated at 4 °C before chemical analysis.

The samples taken from different sampling sites have been analyzed for various heavy metals such as Zinc, Copper, Lead, Cadmium and Iron as per the suggestions given by APHA (1989) [24]. Concentrations of heavy metals in water samples were determined with an atomic absorption spectrophotometer (GCB-Avanta) with a specific lamp for particular metal. Average values of three replicates were taken for each determination. Appropriate drift blank was taken before the analysis of samples. The working wave length for the heavy metals are 248.3 nm for Fe, 213.9 nm for Zn, 324.7 nm for Cu, 228.8 nm for Cd, 217 nm for Pb.

2.3 Multivariate Statistical Analysis

2.3.1 Correlation Analysis Between Heavy Metals

Multivariate statistical analysis can be used to assess the complex eco toxicological processes by showing the relationship and interdependency among the variables and their relative weights. The mathematical models are used to estimate water quality parameters and to describe realistic water situations. Correlation analysis measures the closeness of the relationship between chosen dependent and independent variables. If the correlation coefficient is nearer to +1 or -1, it shows the probability of linear relationship between the variables X and Y. In this study, the relationship of water quality parameters on each other was determined by calculating Karl Pearson's correlation coefficient "r" by using the formula as given.

$$r = \frac{\sum(X-\bar{X})(Y-\bar{Y})}{\sqrt{(X-\bar{X})^2(Y-\bar{Y})^2}} \quad (1)$$

If the values of correlation coefficient "r" between the two variables X and Y are fairly large, it implies that these variables are highly correlated. In such cases it is fissile to try linear relation in the form,

$$Y = a + bX \quad (2)$$

The value of empirical parameters a and b are calculated with the help of the following equation,

$$b = \frac{\sum XY - \bar{X}\sum Y}{\sum X^2 - \bar{X}\sum X} \quad (3)$$

$$a = \bar{Y} - b\bar{X} \quad (4)$$

Where, $\bar{X}$=mean of all values of X $\bar{Y}$= mean of all values of Y

To study the correlation between various water quality parameters, the regression analysis was carried out using computer software SPSS, version-15.

The systematic calculation of correlation coefficient between water quality variables and regression analysis provide indirect means for rapid monitoring of water quality. The correlation coefficient measures the degree of association that exists between two variables, one taken as dependent variable. The greater the value of regression coefficient, the better is the fit and more useful the regression variables. Correlation is the mutual relationship between two variables. Direct correlation exists when increase or decrease in the value of one parameter is associated with a corresponding increase or decrease in the value of other parameter. The correlation between the parameters is characterized as strong, when it is in the range of +0.8 to +1.0 and -0.8 to -1.0,moderate when it is in the range of +0.5 to +0.8 and -0.5 to -0.8,weak when it is in the range of +0.0 and -0.5. The correlation among the different parameters will be true when the value of correlation coefficient (r) is high and approaching to one. These correlation coefficient values can be used in calculating the other parameters of the particular area without analyzing with the help of equation of linearity.

2.3.2 Principal Component Analysis and Factor Analysis (PCA and FA)

Principal Component Analysis and Factor Analysis methods are aimed at finding and interpreting hidden complex and casually determine relationships between features in a data set. The key idea of principal component analysis is to quantify the significance of variables that explain in the observed groupings and patterns of the inherent properties of the individual objects. Factor analysis is a useful tool for extracting latent information, such as not directly observable relationships between variables. The original data matrix is decomposed into the product of a matrix of factor scores plus a residual matrix. In general, by applying the eigen value-one criterion, the number of extracted factors is less than the number of measured features. Factor loadings values of > 0.75, between 0.75 to 0.5 and 0.5 to 0.3 are classified as strong, moderate and weak based on their absolute values. So the dimensionality of the original data space can be decreased by means of factor analysis.

2.3.3 Cluster Analysis (CA)

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 different clusters. In this study, heavy metals classification was performed by the use of cluster analysis (z-transformation of the input data, squared Euclidean distance as similarity measure and Ward's method of linkage) and dendrogram was generated. Since hierarchical agglomerative cluster analysis is used, the number of clusters was also decided by water environment quality, which is mainly affected by land use and industrial structure.

3. Results and Discussion

Zinc is one of the important trace elements that play a vital role in the physiological and metabolic process of many organisms. Nevertheless, at higher concentration, zinc can be toxic to the organisms. As per Indian standards, the desirable limit is 5 mg/L and permissible limit is 15 mg/L [25]. Copper enters the water system through mineral dissolution and industrial effluents, because of its use as insecticide and through corrosion of copper alloy water distribution pipes. According to BIS [25], the desirable limit is 0.05 mg/L and permissible limit is 1.5 mg/L [25]. Lead enters the human body in many ways. It can be inhaled in dust from lead paints or waste gases from leaded gasoline. The desirable limit is 0.05 mg/L as per Indian standards [25]. High concentration of lead in the body can cause death or permanent damage to the central nervous system, the brain and kidneys. According to BIS the desirable limit of Cd is 0.01 mg/L [25]. It replaces zinc biochemically and causes high blood pressures and kidney damage. It interferes with enzymes and causes a painful disease called Itai-itai. Iron is an essential and non-conservative trace element found in significant concentration in drinking water because of its abundance in the earth's crust. The high concentration of iron in ground water is abstracted by drilling water wells both in rural and urban areas for drinking and irrigation purposes. In water, it occurs mainly in ferrous or ferric state. The desirable limit is 0.3 mg/L and permissible limit is 1.0 mg/L as per BIS standards (BIS, 1991). But none of the samples Virudhunagar district (S_1 to S_8) showed the studied metal concentrations above the desirable limit (Fig. 2).

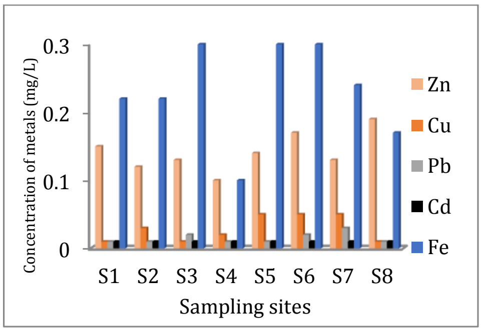

Fig. 2 The concentration of metals at all the sampling sites of Virudhunagar district (S_1 to S_8)

3.1 Correlation Analysis between Heavy Metals

From correlation matrix (Table 2), the highest correlation is observed between lead and copper (r=+0.862) for the 8 sampling sites in Virudhunagar district. The significant correlation between heavy metals indicated the similar source. This correlation could be attributed to the industrial and agricultural discharge as well as combustion of petrol in automobile cars.

Table 2 Pearson's correlation matrix of heavy metals in the groundwater in Virudhunagar district

	Zn	Cu	Pb	Cd	Fe
Zn	1				
Cu	0.137	1			
Pb	0.235	0.862	1		
Cd	0.358	0.324	0.422	1	
Fe	0.296	0.453	0.311	0.399	1

3.2 Principal Component Analysis and Factor Analysis

Principal Component Analysis (PCA) with varimax rotation was applied to the water quality data set to form a correlation matrix for 8 sampling sites in Virudhunagar district and assist in the identification of sources of metal pollution. The percentage of variance by different components extracted is displayed and the factor loadings of the different variables have been presented in Table 3. Two factors were obtained on metals displaying a cumulative variance of 75.04% (Table 3).

Factor 1 accounts for 44.63% of the total variance and is dominated by the strong factor loadings for Cu (0.874) and other metals Pb, Cd, Fe and Zn demonstrate low factor loadings in this case indicating their independency within this group. This factor is assumed to originate from natural and anthropogenic contribution.

Factor 2 accounts for 30.41% of the variance and is dominated by Pb (0.810) having positive strong factor loadings. This factor is mainly due to pronounced anthropogenic inputs and weathering process.

Table 3 Total variance explained and component matrixes for heavy metal contents of Virudhunagar district

Element	Communality	Eigen value	Total variance	Cumulative total variance	Factor 1	Factor 2
Zn	1.000	2.231	44.63	44.63	0.170	-0.394
Cu	1.000	1.521	30.41	75.04	0.874	0.245
Pb	1.000	0.631	12.63	87.67	0.362	0.810
Cd	1.000	0.514	10.27	97.94	0.126	0.475
Fe	1.000	0.103	2.058	100.00	0.434	-0.147

3.3 Cluster Analysis of 8 Sampling Sites of Virudhunagar District

The most similar objects are first grouped and these initial groups are merged according to their similarities. Eventually as the similarity decreases all subgroups are merged into a single cluster. The cluster analysis of the heavy metals in 8 sampling sites of Virudhunagar district showed 3 clusters (Fig. 3). Cluster I consists of Pb-Cu, Cluster II and cluster III are distinct clusters which shows their independent sources. Cluster I is mainly due to mineral dissolution and industrial effluents. Cluster II occurred due to the natural and anthropogenic sources in the environment. Cluster III suggested that from geogenic sources and the use of liquid manures.

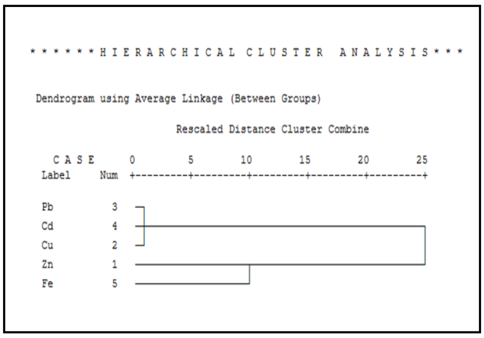

Fig. 3 Dendrogram showing clustering of trace elements in Virudhunagar district (S_1 to S_8)

4. Conclusion

Principal component analysis and cluster analysis, in combination with metal concentration analysis and correlation analysis can be effective tools for the characterization of the sources of the pollutants. The Cluster analysis organized the metals into groups such that within-group similarity is maximized and among-group similarity is minimized. This analysis revealed that Fe showed almost independent pattern among the other studied metals. Variation of Zn, Cu, Pb, and Cd concentrations is controlled by anthropogenic intense agriculture activities. The concentration of Fe is mainly affected by natural parent material and human activities. Therefore, multivariate statistics is found to be a powerful tool to identify the main factors determining the variability of geochemical data and interpret the measurement results. It is used to design a future spatial sampling strategy and reduced water variable examinations in an optimal monitoring network.

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