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ASSESSMENT ON METAL ELEMENTS IN SOIL OF WASTE LANDFILL AT KHULNA: A STUDY BASED ON MULTIVARIATE STATISTICS, GIS AND BP-ANN

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ABSTRACT

The main focus of this study was to assess the spatial distribution of metal elements presence in soil of waste disposal site at Rajbandh, Khulna, Bangladesh. Sixty soil samples were collected at a depth of 0-30 cm from the existing ground surface from the selected waste disposal site, forty samples in dry and twenty samples in rainy season. In the laboratory, the relevant parameters of Fe, Mn, Cr, Cu, Pb, Zn, Ni, Cd, As, Hg, Co Na, K, Ca, Al, Ti, Sb, Sc, Sr, V and Ba in soil were measured through the standard methods. The conventional statistics were performed by SPSS analysis. Result reveals that all the measured metals were not normally distributed, showing positively skewed data except Fe. Furthermore, Person's correlation was also performed using SPSS which represents the high significant positive relationship between metal elements indicating close association with each other. Principal component analysis (PCA) was performed using XLSTAT and it reveals that most of the metal elements derived from anthropogenic activities. In this study, ordinary kriging (OK) and inverse distance weighting (IDW) with power of 1-5 were used to evaluate spatially dependence of metal elements. The semi variogram showed that the metal elements of Fe, Zn, Cd, Hg, Co, K, Ca and Tiin soil exhibited strong spatial correlation. Furthermore, Mn, Cr, Na and Ba were found moderate spatially dependent and Ni, As, Sb were weakly spatially correlated. From ANN analysis through MATLAB, regression coefficient, R was found very closer to 1 almost for every metal element in the dry season compared to rainy season, which indicates accuracy of observed and predicted data obtained from ANN. In addition, performance assessments of interpolation methods were done on the basis of mean absolute percentage error (MAPE), G- value and relative improvement (RI). Results indicated that IDW1 was most accurate performance in all applied assessment methods. Based on the results, it is evident a systematic and constant monitoring for metal elements pollution should be introduced and certain remediation steps should be taken to minimize the rate and extent pollution problems in future.

Keywords: *Artificial neural network, Metal elements, Pearson's correlation, Principal component analysis, Waste disposal site, Inverse distance weighting.*

1. INTRODUCTION

The effects of metal elements are found to vary with the conditions prevailing in the dumping sites and its binding forms (Pebesma *et al.,* 2007). In addition, the contamination of soil with metal elements draws prodigious consideration due to its non-biodegradable features impending threat to food safety and injurious belongings on the environmental components. There has long been concern about the issue of contamination of soil by metal elements because of their toxicity for plant, animal and human beings as well as their lack of biodegradability (Zhuang *et al.,* 2009). Interests arise among the researchers to find out the origin and consequences of these metal elements on the earth due to its toxic and detrimental effects in the environment (Jia *et al.,* 2010).The applications of multivariate statistical approaches permit a better technique for classification, modeling and interpretation of soil monitoring data (Smith *et al.,* 2007). In addition, spatial distribution is essential for assessing the effect of metal elements in soil and to delineate contamination zones (Omran and Razek, 2012). Deterministic interpolation approaches, such as inverse distance weighting (IDW) method as well as Radial basis functions (RBF) and Local polynomial interpolation (LPI) through ArcGIS are also used to predict of values, standard error and condition number that are comparable to ordinary kriging with measurement errors (Zhang, 2011). A study of Creutin and Obled (1982) compared the performance of several interpolation techniques from produced prediction surfaces. Furthermore, the application of SOM-ANN is useful for recognizing spatial patterns in contaminated zones as well as water quality assessment for pollutant sources identification, pattern recognition and classification (Lee *et al*., 2006).

Khulna is the third largest metropolitan city among ten metropolitan cities of Bangladesh. Total rate of MSW generation in Khulna is found to be 420 to 520 ton per day, directly disposed at Rajbandh waste disposal site which is the only official dumping site over 25 acres in area (Murtaza, 2012) . The waste disposal site is 10 kilometers far from City Corporation headquarter in the direction of west. Unplanned and uncontrolled MSW disposal arise the necessity to carry out an intensive study of possible generation sources of such pollution on in and around the disposal site. To these endeavors, total sixty soil samples were collected at a depth of 0-30 cm from the existing ground surface and the relevant metals elements of Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mn, Na, Ni, Pb, Sb,

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Sc, Sr, Ti, V and Zn was measured through the standard test methods in the laboratory. In this study, conventional statistical analysis including normality test and descriptive tests were performed using SPSS to analyze the distribution and variation of metal concentration in soil of waste disposal site. Multivariate statistics such as Pearson's correlation and principal component analysis (PCA) were performed using XLSTAT for finding the generation sources of metal elements in soil. In addition, the spatial distributions of the concentration of metal elements in soil were performed using ordinary kriging (OK) and inverse distance weighting with power of 1-5 interpolation technique by ArcGIS. In addition, semivariogram parameters obtained from Geostatistics exhibited spatial dependence of metal elements in soil. Performance evaluation of the interpolation methods was evaluated by defining the error using existing literature indices. Artificial neural network (ANN) was performed to predict the concentration of metal elements as well as the accuracy of predicted concentration of metal elements using MATLAB software. The objectives of this study are: (i) to show the temporal pattern and distribution of metal concentration; (ii) to evaluate the correlation of metal elements and their possible sources of generation; (iii) to show the spatial extent of concentration of metal elements in and around the soil of waste disposal site; (iv) to check the accuracy of predicted metal concentration obtained from ANN.

2. MATERIALS AND METHOD

The overall research procedure and materials utilized in this study are described and hence discussed in the following articles.

2.1 Study Area

Khulna is the third established metropolitan city located in the Khulna division of Bangladesh. It covers an area of 4394.45 km² and is bordered on the north by the Jessore district and the Narail district, on the south by the Bay of Bengal, on the east by the Bagerhat District, and on the west by the Satkhira district. The geological location of Khulna is 22.35^0N and 89.30^0E. Urban development results a huge amount of waste generation poses a great thread to the environment and human health. The selected waste disposal site, Rajbandh is the only certified waste dumping site of Khulna. Based on aforementioned authenticities, it has become inevitable of comprehensive study of possible generation sources of metal elements in soils to minimize their spread over the study area.

In total sixty soil samples, forty samples were collected in dry season (March to May 2016) then rest twenty samples were collected in rainy season (June to August 2016). In dry season, the first sampling point referred as BH-1 was at the center of waste disposal site followed by a gradual addition of about 10 m distance by the subsequent boreholes. On the other hand, the first sampling location of rainy season referred as BH-41 is about 30 m apart from BH-1 maintaining a gradual addition of about 15 m in selecting other following boreholes. Figure 1 depicted the soil sampling locations in waste disposal site at Rajbandh, red circles indicated sampling points in dry season and blue triangles indicated sampling points in rainy season.

2.2 Laboratory Investigation

Concentration of relevant metal elements in soil was measured maintaining standard procedure in laboratory. At first 10 g of each soil sample was taken into a 100 mL conical flask washed with deionized water and left overnight. Each sample was kept into the temperature of 150°C for about 90 minutes followed by raising the temperature up to 230°C for 30 minutes. Subsequently, re-digestion of samples was done by adding HCl solution in ratio of 1:1 for another 30 minutes. The mixture obtained was cooled down to room temperature. After performing the digestion procedure, metal element concentrations in this digested solution were determined using atomic absorption spectrophotometer (AAS) in the laboratory and the amount of each metal element was deduced from the calibration graph and reported in mg/kg.

2.3 Pearson's Correlation Coefficient

Accumulated concentrations of metal elements irrespective to their sources can be computed by Pearson's correlation coefficient, r provided in Equation (1). High correlation coefficient values reflect that the accumulated concentrations of metal elements come from similar pollution sources (Li *et al.,* 2012).

$$r = \frac{\sum_{i=1}^{n}(x_i-\bar{x})\,(y_i-\bar{y})}{\sqrt{\sum_{i=1}^{n}(x_i-\bar{x})^2}\sqrt{\sum_{i=1}^{n}(y_i-\bar{y})^2}} \tag{1}$$

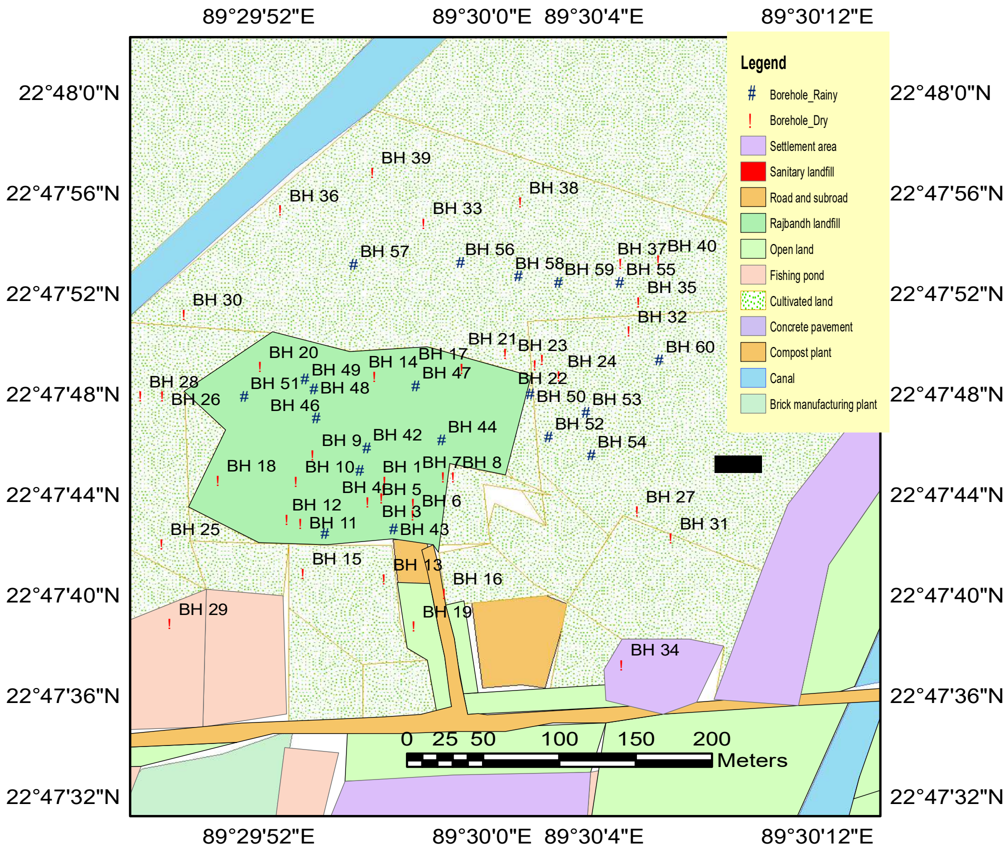

Figure1: Map showing of soil sampling locations in waste disposal site

2.4 Principal Component Analysis

In this study, the PCA method was performed using following Equation 2to determine the directions of which the input variables display the most substantial variability.

$$\mathrm{PC1} = a_1 x_1 + a_2 x_2 + \cdots + a_n x_n$$

$$\mathrm{PCn} = \sum_{j=1}^{n} a_{1j} x_j \qquad (2)$$

Where, a_{1j}= eigenvectors obtained from the correlation matrix and x_j= input variables

Factor loadings lead to identification of possible generation sources of pollution by metal elements. It seems reasonable to tentatively identify the first rotated factor as “anthropogenic activities” and second rotated factor as “natural sources”. Moreover, some previous investigations indicated first principal component (PC1) and second component (PC2) refers to the contamination of soil due to anthropogenic or human activities and natural parent materials, respectively (Tahir *et al.,* 2007).

2.5 Ordinary Kriging

Ordinary kriging (OK) delivers estimation at an unobserved location of variable z, based on the weighted average of adjacent observed sites within a given area, is defined by the following equation (Yasrebi *et al.,* 2009):

$$Z(s_0) = \sum_{i=1}^{n} \lambda_i Z(s_i) \qquad (3)$$

The ordinary kriging (OK) with eleven distinct models such as Circular, Spherical, Tetraspherical, Pentaspherical, Exponential, Gaussian, Rational quadratic, Hole effect, K-Bessel, J-Bessel and Stable was performed to select the best fitted model for each metal elements in soil. This selected model was used for modeling the empirical semivariogram and spatial distribution of the concentration of metal elements in soil. A study conducted by Brovelli *et al.* (2011) and stated when MSPE closed to 0 and RMSSPE tends to 1 for a particular model, then the model will be chosen as the best fitted model. In addition, when ASPE closed to RMSPE for a specific model, then it can be confidently said that the prediction model was appropriate (Johnston *et al*., 2001). Using the model semivariogram, basic spatial parameters such as nugget variance (Co), range (A) and sill (C + Co) were calculated. Nugget variance is defined as the variance at nil distance, sill and range is defined as the lag distance between measurements at which one value for a variable does not influence neighboring values and the distance at which values of one variable become spatially independent of another respectively (Lopez-Granados *et al.,* 2002). Spatial dependence for the soil variables were assessed by the ratio between the nugget semivariance and the total semivariance (Cambardella, 1994).

2.6 Inverse Distance Weighting

In inverse distance weighting (IDW) method, it is expected substantially that the rate of correlations and similarities between neighbors is proportional to the distance between them which is defined as a distance reverse function of every point from neighboring points. The important factor of inverse distance interpolatoris the value of the power parameter (Isaake and Srivastava, 1989). In this study, estimations were made using different integer powers of 1 to 5 using the equation (4) (Kravchenko and Bullock, 1999). The lower the root mean square prediction error (RMSPE), the better the interpolation technique.

$$Z_0 = \frac{\sum_{i=1}^{N} z_i {d_i}^{-n}}{\sum_{i=1}^{n} {d_i}^{-n}} \tag{4}$$

where, Z_0 is the estimation value of variable z in point I, Z_i is the sample value in point I, d_i is the distance of sample point to estimated point, N is the coefficient that determines weigh based on a distance, n is the total number of predictions for each validation case.

2.7 Assessment of Method Performance

The precision of prediction method was measured by Mean Absolute Percentage Error (MAPE), which is the percentage error between predicted and observed value (Yao *et al*., 2013). Small values of MAPE represent a model with less errors and more accurate predictions (Yao *et al*., 2013). The MAPE is computed by the Equation (5) given below (Yao *et al.,* 2013; Yasrebi *et al.,* 2009):

$$MAPE = \frac{1}{n}\sum_{i=1}^{n}[(p_i - o_i)/\, o_i] \tag{5}$$

The effectiveness of the models was evaluated using a goodness of prediction statistic (G), also known as the coefficient of determination, is computed using Equation (6) (Yao *et al.,* 2013; Yasrebi *et al*., 2009):

$$G = 1 - [\frac{\sum_{i=1}^{n}(p_i - o_i)^2}{\sum_{i=1}^{n}(o_i - \bar{o})^2}] \tag{6}$$

Where n is the number of validation points, p_i is the predicted value at point i, o_i is the observed value at point i, and $\bar{o}$ is the sample arithmetic mean. A G-value equal to 1 indicates perfect prediction, a positive value indicates a more reliable model than if the sample mean had been used, a negative value specifies a less reliable model than if the sample mean had been used, and a value of zero indicates that the sample mean should be used (Yao *et al*., 2013).

Finally, the relative improvement (RI) of the best method compared with the others is calculated with equation (7) (Yasrebi *et al*., 2009):

$$RI = \frac{100\,|RMSE_{Best} - RM \quad Current|}{RMSE_{Best}} \tag{7}$$

2.8 Back–Propagation (BP) Network

In this study, known as the sampling points, will undergo training and testing the BP network model for spatial interpolation, the interpolation function is net.trainFcn = 'trainlm', [net,tr] = train(net,...) (Rumelhart *et al*.,1986). In order to test the accuracy of the network, the network output and the actual measured value after standardization for regression analysis for both the dry and rainy seasons, respectively, where, R is the output value and the correlation coefficient between the actual value, can be seen from the figure the network with very good generalization performance (Rumelhart *et al*., 1986).

3. RESULTS AND DISCUSSION

3.1 Descriptive Statistics

The descriptive statistics analysis was performed for 40 soil samples collected from waste disposal site in the dry season to analyze the different metal elements parameters are summarized in Table 1. For all metals, the coefficients of variation (CV) varies from 22.11% of Zn to 59.41% of Cu in dry season and 18.25% of Zn to

77.25% of Mn in rainy season respectively, which indicate a great degree of variability. Non-homogeneous distribution of concentrations of emitted metal elements due to anthropogenic activities results elevated degree of variation. The greatest and the smallest standard deviations were detected for Ti (406.2571, 297.8519) and Cd (1.137265, 0.736449) in dry season and rainy season, respectively. Large standard deviations were found in all metal elements, especially in case of Fe, Al, K and Ca in both the dry and rainy seasons. This also indicated the wide variation of concentrations in soil of waste disposal. The concentrations of all metal elements were not normally distributed, showing positively skewed data in both the dry and rainy seasons. There was a remarkable change in the content of metal elements among the sampled soils. The concentration of Fe and Ti were the most significant in both the dry and rainy season, with the concentration of Al>K >Ca >Ba >Na >Pb >V >Sr >Zn >Mn >Sc >Cu >Sb >Co >Cr >Hg >As >Ni >Cd in dry season and Al>K >Ca>Ba >Na >V>Sr>Zn > *Pb* >Mn >Sc >Co >Cu >Sb >Cr >Hg >As >Ni >Cd in rainy season. The concentrations of metal elements were relatively lower as compared to the dry season and magnitude of concentrations follow almost same pattern during both the seasons.

Table 1: Descriptive statistics of soil parameters (0-30 cm depth) of 40 samples in dry season

Metal elements	Minimum	Maximum	Mean	Median	SD	CV (%)	Skewness	Kurtosis
Fe	733.2	1987.7	1363.9	1386.5	350.15	25.67	0.81	-1.20
Mn	10.82	30.76	15.692	14.155	4.68	29.82	1.13	2.88
Cr	4.19	9.82	6.0341	5.83	1.43	23.63	1.10	0.14
Cu	2.92	16.45	6.2026	4.815	3.68	59.41	1.55	1.43
Pb	21.30	90.55	37.608	33.935	13.67	36.34	1.00	5.16
Zn	22.79	50.76	34.569	34.635	7.65	22.11	1.15	0.03
Ni	2.56	8.06	4.8346	4.7075	1.60	33.02	0.70	-0.95
Cd	2.55	7.03	4.55	4.46	1.14	24.99	0.60	-0.44
As	1.55	8.77	4.1517	3.415	2.03	48.79	1.27	-0.64

Table 2: Correlation analysis and coefficients for the metal elements

	Fe	Mn	Cr	Cu	Pb	Zn	Ni	Cd	As
Fe	1								
Mn	0.844	1							
Cr	0.923	0.827	1						
Cu	0.769	0.791	0.717	1					
Pb	0.847	0.895	0.770	0.851	1				
Zn	0.940	0.87	0.880	0.862	0.879	1			
Ni	0.894	0.81	0.835	0.890	0.837	0.897	1		
Cd	0.949	0.892	0.906	0.830	0.890	0.911	0.911	1	
As	0.885	0.794	0.820	0.905	0.874	0.89	0.951	0.910	1

3.2 Correlation of Metal Elements in Soil

The concentration of all parameters showed a high significant positive relationship with each other, such as K showed a significant positive relationship with of Fe (0.936), Mn (0.805), Cr(0.805) , Cu(0.871), Pb (0.842), Zn (0.901), Ni (0.929) , Cd (0.925), As (0.921) , Hg (0.856) , Co (0.86) , Na (0.969) Additionally, the correlations between Ca, V and Fe, Mn, Cr, Cu, Pb, Zn, Ni, Cd, As, Hg, Co, Na, K, Al, Ti, Sb, Sc, Sr, were significantly positive (Table 2). The most significant correlation was observed for Sb and Sr (0.988), Sc and Sr (0.986), V and Sr (0.986), Al and Ca (0.984), Ti and K (0.963), Ni and Ti (0.965), K and Na (0.969), Co and Ba (0.938), Al and As (0.976) and Fe and Cd (0.949). Such significant correlations between metals may reflect that these metal elements had similar pollution level and similar pollution sources. The lowest correlation coefficient lies between Cr and Co (0.7). However, the concentrations of Cr showed very weak correlations with Cd and Co relative to others. This indicated that Cr was from different sources than Cd and Co. Table 2 shows the correlation between some of metal elements in dry season.

3.3 Principal Component Analysis

For variability calculation based on eigenvector and factor loadings, PCs of 21 for dry and 19 for rainy were considered for the metal elements and results provided in Table 3. The larger eigenvalue obtained for F1 (18.267) indicated large proportion of variability (86.987%) for dry season (Table 3). Based on the results of PCA for metal elements of dry season, the eigenvalues up to the second extracted components (F2) were found greater than 1.0. Moreover, as the eigenvalues for the PCs (F3 to F21) as well as (F2 to F19) were found less than 1 for dry and rainy season, respectively, so, these PCs were depicted as very negligible contribution in the analysis. Thus, the variables could be reduced to 2 components model (dry season) with 92.105% variation as well as 1 component model (rainy season) that accounts for 88.253% variation.

Table 3: PCA of metal elements in soil for dry and rainy seasons

PCs	Dry season			Rainy season		
	Eigenvalue	Variability	Cumulative	Eigenvalue	Variability	Cumulative
F1	18.267	86.987	86.987	18.533	88.253	88.253
F2	1.070	5.118	92.105	0.906	4.3147	92.567

Varimax rotation was applied to simplify the factor interpretation by reducing total number of variables to distribute the importance more or less evenly in the factor space. Moreover, some previous investigations indicated first principal component (PC1) and second component (PC2) refers to the contamination of soil due to anthropogenic or human activities and natural parent materials, respectively (Tahir *et al.,* 2007).

Table 4: Factor analysis of PC's before and after varimax rotation for dry season

Metal	Before rotation					After rotation	
	F1	F2	F3	F4	F5	D1	D2
Fe	0.96	-0.24	0.02	-0.11	0.01	0.91	0.37
Mn	0.72	0.61	0.29	-0.07	0.00	0.22	0.91
Cr	0.78	-0.28	0.32	0.44	-0.08	0.79	0.24
Cu	0.89	0.28	-0.11	0.25	0.09	0.55	0.75
Pb	0.85	0.40	0.18	-0.14	0.00	0.45	0.82
Zn	0.92	0.09	0.02	0.10	0.35	0.68	0.62
As	0.97	0.01	-0.15	0.05	-0.07	0.77	0.59

Based on the results of PCA for metal elements in dry season provided in Table 4, it can be decided that the metal elements of Cu, Hg, Mn, Pb, and Zn derived from natural sources as they were closed to PC2, whereas, Al, As, Ba, Ca, Cd, Co, Fe, K, Na, Ni, Sb, Sc, Sr, Ti and V were also derived from anthropogenic activities because they were closed to PC1. Nevertheless, the metal element of Cr also derived from both natural soil parent materials and anthropogenic sources (Table 4). In context of rainy season, the PCA results demonstrated that metal elements of As, Cr, Hg, K, Mn and Na were derived from different sources as the location of these metal elements were distinct from other metal elements of Al, Ba, Ca, Co, Cu, Fe, Ni, Sb, Sc, Sr, Ti and V. Moreover, it can be estimated as the metal elements of Al, Ba, Ca, Co, Cu, Fe, Ni, Sb, Sc, Sr, Ti and V were derived from anthropogenic activities and As, Cr, Hg, K, Na and Mn were derived from natural parent materials. Other metal elements of Cd, Pb and Zn were derived from both natural parent materials and anthropogenic sources.

3.4 Spatial distribution of metal elements

The spatial distribution of metal concentrations is a convenient tool to identify the sources of generation of metal elements as well as contamination hotspots with high metal concentrations in a visual form. The predicted map of almost all the metal elements showed almost similar pattern of contamination indicated same sources of generation indicating anthropogenic activities accompanied by particular natural soil materials.

3.4.1 Inverse Distance Weighting

In this study, cadmium (Cd) was taken for instance. IDW with power 5 (IDW5) exhibited the lowest RMSPE value of 1.324 indicated the lowest prediction error in case of Cd (Table 5).

Table 5: Cross validation of IDW for Cd

Metal element	Power	[a]MPE	[b]RMSPE
Cd	1	0.675	1.445
	2	0.594	1.359
	3	0.495	1.324
	4	0.412	1.318
	5	0.349	1.306

[a]MPE=Mean Prediction Error, [b]RMSPE= Root Mean Square Prediction Error

Table 6: Cross validation results of ordinary kriging for different models of Cd in soil

Models	[a]MPE	[b]RMSPE	[c]ASPE	[d]MSPE	[e]RMSSPE
Circular	0.0244	1.2724	1.4398	0.0081	0.9097
Spherical	0.0361	1.2590	1.4399	0.0161	0.8994
Tetraspherical	0.0436	1.2520	1.4348	0.0199	0.8986
Pentaspherical	0.0442	1.2475	1.4141	0.0200	0.9148
Exponential	0.0597	1.2521	1.4073	0.0245	0.9244
Gaussian	0.0295	1.2638	1.4375	0.0107	0.9102
Rational quadratic	0.0959	1.2498	1.3924	0.0358	1.0139
Hole Effect	0.0524	1.2634	1.4322	0.0310	0.9219
K-Bessel	0.0550	1.2359	1.4111	0.0231	0.9043
J-Bessel	0.0220	1.2359	1.4284	0.0087	0.8980
Stable	0.0630	1.2322	1.4074	0.0275	0.9036

[a]MPE= Mean Prediction Error, [b]RMSPE= Root Mean Square Prediction Error, [c]MSPE= Mean Standardized Prediction Error, [d]RMSSPE= Root Mean Square Standard Prediction Error, [e]ASPE= Average Standard Prediction Error.

Table 7: Fitted parameters of the theoretical variogram model for metal elements parameters

Metals	Models	Predicted error				
		[a]MPE	[b]RMSP	[c]ASPE	[d]MSPE	[e]RMSS
Fe	J-Bessel	17.891	496.523	482.812	0.00085	1.0234
Mn	J-Bessel	0.0597	5.627	6.737	-0.0032	0.880
Cr	Circular	-0.0038	2.081	2.217	-0.0045	0.965
Cu	Hole Effect	0.1637	2.5916	3.5444	0.04146	0.7525
Pb	Hole Effect	-0.516	12.1973	17.4139	-	0.7203
Zn	Hole Effect	-0.0196	8.0749	9.1687	-0.0402	0.9258
Ni	Hole Effect	0.0938	1.3416	1.4173	0.0484	0.9767
Cd	Rational	0.0958	1.2498	1.39247	0.03582	1.0139
As	J-Bessel	0.0701	1.4997	1.8929	0.0269	0.8365

[a]MPE= Mean Prediction Error, [b]RMSPE= Root Mean Square Prediction Error, [c]MSPE= Mean Standardized Prediction Error, [d]RMSSPE= Root Mean Square Standard Prediction Error, [e]ASPE= Average Standard Prediction Error.

3.4.2 Ordinary Kriging

The cross-validation result of eleven distinct for Cd is shown in Table 6. The values of MSPE range from 0.0081 to 0.0358 and RMSSPE from 0.8980 to 1.0139. Result reveals the value of MSPE was closest to zero (0.0358), RMSSPE closest to 1(1.0139) and the ASPE (1.3924) closed to RMSPE (1.2498) for the model of rational quadratic (Table 6). So, in this case, rational quadratic was selected as the best fitted model using ordinary kriging interpolation. In addition, the best fitted model from cross validation results of eleven distinct models using ordinary kriging interpolation for the studied metal elements of Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mn, Ni, Pb, Sb, Sc, Sr, Ti, V and Zn in soil is provided in Table 7.

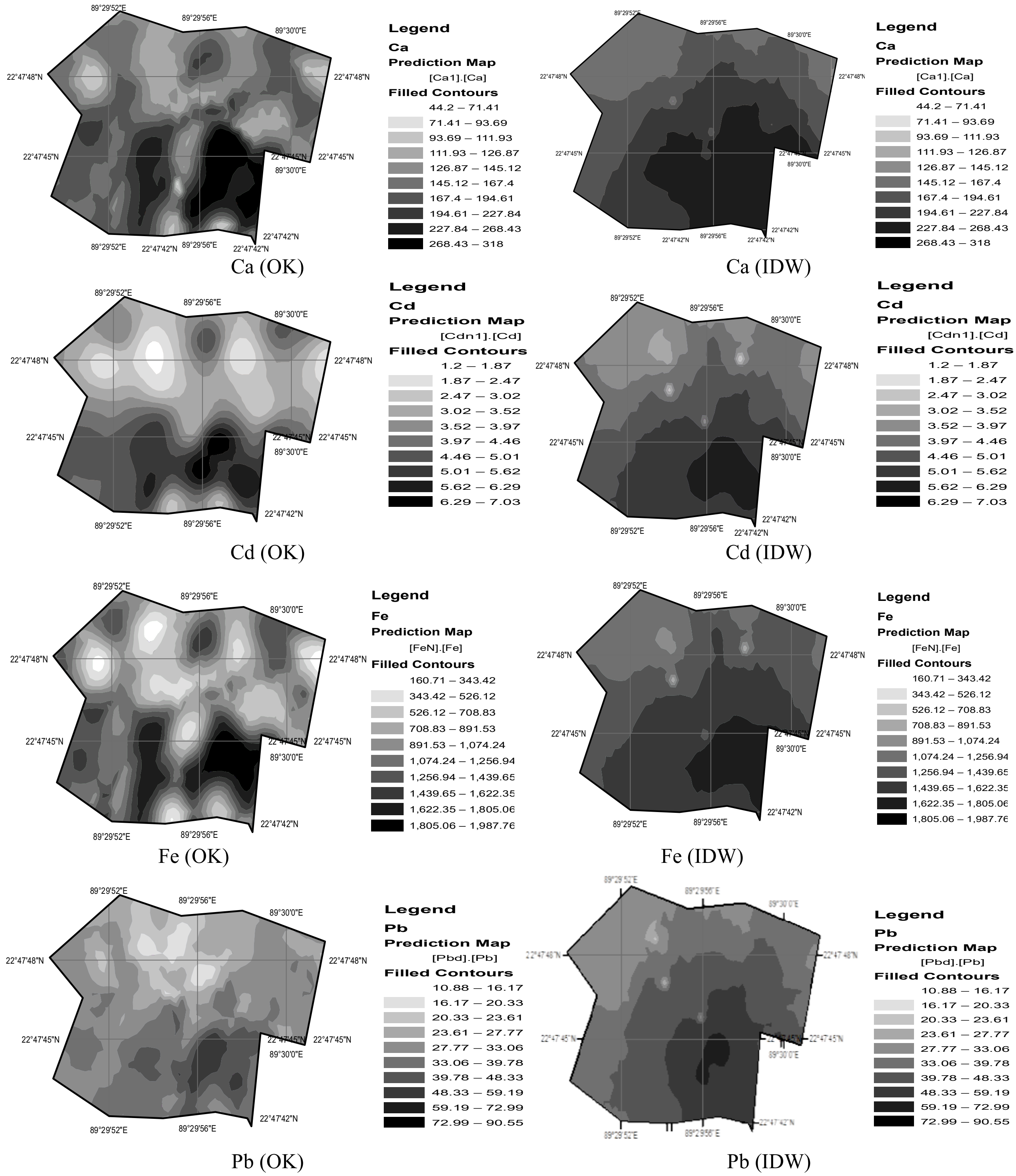

Figure 2: Surface spatial distribution of metal elements in soil using OK and IDW1

The semivariagram parameters such as range, nugget, partial sill and nugget to sill ratio from obtained best fitted model for some of studied metal element are provided in Table 8. A study conducted by Yasrebi *et al*. (2010) and stated that when the nugget to sill ratio, $C_0/(C_0 + C)$ was found <25%, 25 to 75% and >75%, it was considered the soil parameters are strongly, moderately and weakly distributed spatially, respectively. In this study, from semivariogram parameters, $C_0/(C_0 + C)$ were found to be 0.099, 8.48, 0.21 and 16.28% for the metal elements of Cd, Co, K, and Ti respectively. This statement (less than 25%) that exhibited that these metal elements were strongly distributed spatially. In addition, $C_0/(C_0 + C)$ were found as 55.44, 47.93, 36.34, 36.34, 74.11, 36.91, 27.96, 46.34, 55.38, 43.90, 39.07, 51.50 and 61.63% for Al, As, Ba, Cr, Cu, Mn, Na, Ni, Pb, Sb, Sc, Sr and V, respectively, indicated that these metal elements in soil were moderately correlated spatially. In contrast, the metal elements of

Ca, Fe, Hg and Zn were non spatially correlated as the C_0/ (C_0 + C) was found to be zero. Figure 2 shows the spatial distribution of some studied metal elements using IDW and OK interpolation techniques.

3.5 Performance of the Interpolation Methods

Table 8 shows the performance assessment of interpolation methods for all studied metal elements in this research. On the basis of MAPE and RMSE, IDW prefers better than OK. In this study, IDW of power 1-5 exhibits value of zero for each parameter which means in IDW as percentage of relative error is zero. In case of G-value, IDW showed value of 1 for each parameter that indicated perfect predictions (Yao *et al.,* 2013). OK showed positive values which indicated more reliable model, but in this case, IDW contributed best result. The Relative improvement (RI) of interpolation techniques is also showed in Table 9. IDW procedure with power of 1 result in significant reduction of RI (0% for all parameters in dry season and rainy season respectively) compared to other IDW powers and OK. Results indicated that IDW with power of 1 was most accurate performance in all applied methods.

Table 8: Semivariogram parameters of best fitted models for metal elements

etals	Models	Range (A)	Nugget (C_0)	Sill (C+C_0)	C_0 / (C+C_0)
Cd	Rational quadratic	0.00124	0.0027087	2.7114126	0.000999
Co	K-Bessel	0.00346	0.8716488	10.277674	0.0848099
Cr	Circular	0.00126	2.2888501	6.2986337	0.3633883
Cu	Hole Effect	0.00358	10.793073	14.564143	0.7410716
Fe	J-Bessel	0.000681	0	269384	0
Hg	J-Bessel	0.00103	0	5.1107591	0
K	Exponential	0.00279	37.111943	17528.732	0.0021172
Mn	J-Bessel	0.000615	17.641389	47.80017	0.3690654
Zn	Hole Effect	0.000644	0	96.449061	0

3.6 Spatial Interpolation based on BP network model

The regression coefficient of Cr for dry and rainy seasons is shown in Figure 3 and Figure 4, respectively. Each figure contains R value for training, validation, test and resultant R value obtained from training, validation and test results. From Figures, it can be observed that R was very close to 1 for dry season, whereas the R value is comparatively outlying from 1 during the rainy season, indicated that the prediction made by BP-ANN for dry season was more accurate than that of rainy season.

Table 9: Performance assessment of interpolation methods

Metals	MAPE		RI						G-value		RMSE	
	OK	IDW (1-5)	OK	IDW 1	IDW 2	IDW 3	IDW 4	IDW 5	OK	IDW (1-5)	OK	IDW (1-5)
Fe	0	0	12.86	0	4.90	7.71	9.39	10.59	1	1	0	0
Mn	0.28	0	19.14	0	5.21	7.76	9.55	11.03	0.86	1	2.50	0
Cr	0.21	0	12.21	0	5.36	8.63	9.90	10.21	0.81	1	1.04	0
Cu	0.29	0	32.15	0	8.59	14.4	18.5	20.77	0.55	1	2.36	0
Pb	0.06	0	21.90	0	6.80	10.5	13.1	15.08	0.82	1	6.24	0
Zn	0	0	15.69	0	5.03	7.41	8.72	9.233	1	1	0	0
Ni	0.10	0	9.154	0	5.90	8.33	9.27	9.282	0.72	1	0.93	0
Cd	0	0	13.47	0	5.90	8.34	8.75	8.344	1	1	0	0
As	0.22	0	23.05	0	10.7	17.9	21.2	22.51	0.42	1	1.18	0

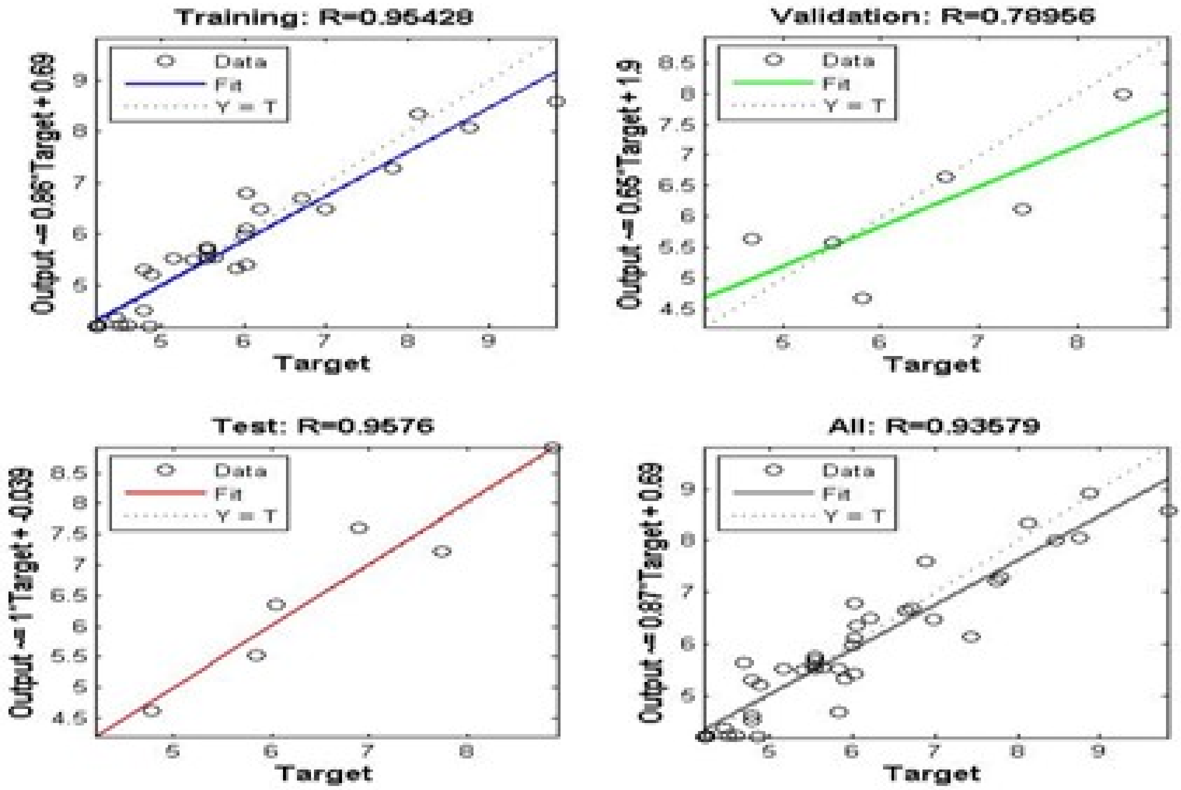

Figure 3: Regression Coefficient of Cr in soil for dry season.

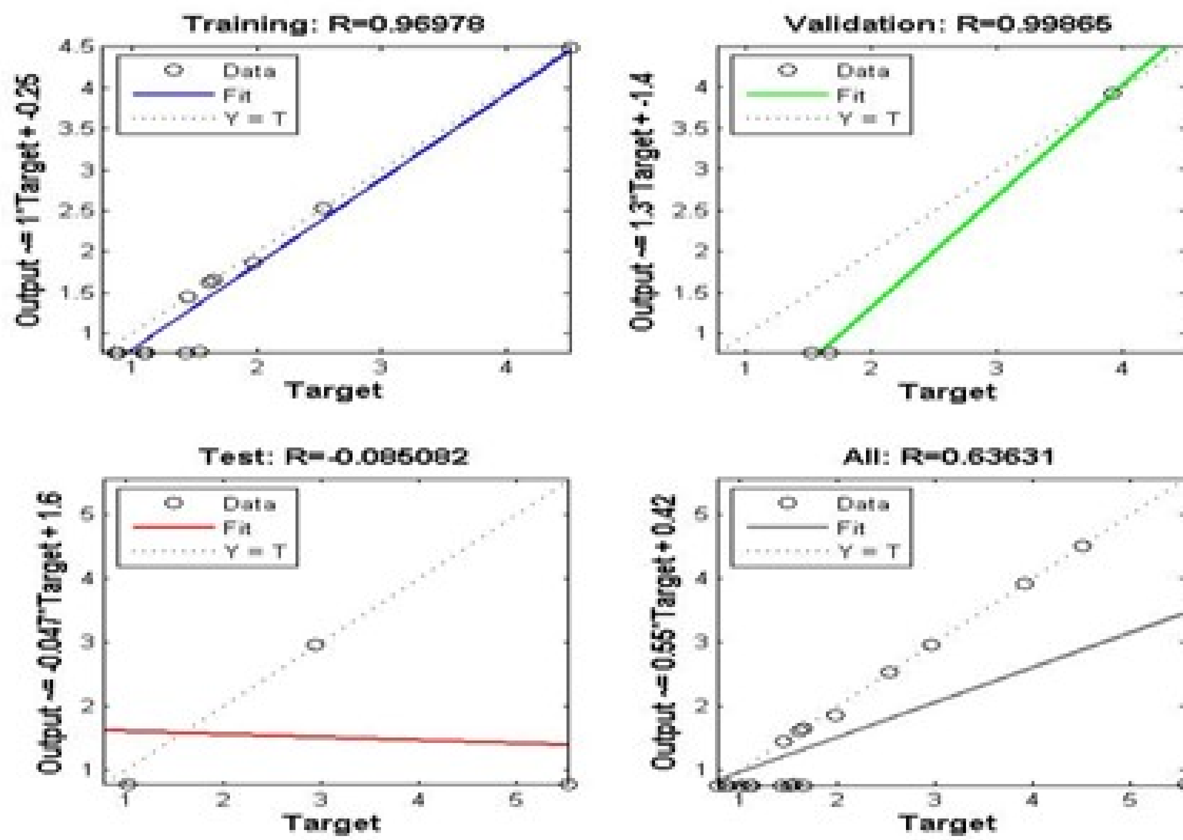

Figure 4: Regression Coefficient of Cr in soil for rainy season.

4. CONCLUSIONS

The concentrations of metal elements in soil was to be found comparatively lower in case of rainy season than that of dry season as well as the magnitude of concentration showed almost the same pattern during both the dry and rainy seasons. The results of Person's correlation represent the association of metal elements with each other irrespective to their pollution sources. The most significant correlation was observed for Sb and Sr, Sc and Sr, V and Sr, Al and Ca, Al and V, Ti and K, Ni and Ti, K and Na, Al and As and Fe which reflect that these metal elements had similar pollution level where the concentrations of Cr showed very weak correlations with Cd and Co indicated that Cr was from different sources than Cd and Co. PCA suggested that Fe, Zn, Ni, Cd, As, Co, K, Ca, Al, Ti, Sb, Sc, Sr, V and Ba were mainly from natural sources and Mn and Pb mainly generated from anthropogenic sources. The geostatistical analysis of OK and IDW (power 1-5) were used to interpolate the metal elements concentration. In addition, the performances of interpolation methods were assessed on the basis of MAPE, RMSE, G-value and Relative Improvement (RI). It is obtained that IDW 1 performed best for each assessment. The ANN was also performed to check the validity and accuracy of the metal elements concentration obtained from laboratory. Therefore, concerns about contamination of metal elements and their possible sources should be taken into account for the betterment of localities in and around waste disposal site.

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