

Estimation of Zn Bonds Using Multi-Layer Perceptron (MLP) Artificial Neural Network Method in Chahnimeh, Zabol

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ABSTRACT

Aims Artificial Neural Networks (ANNs) are powerful tools that are commonly used today in prediction deposit-related sciences. The research aimed at predicting various five links of heavy metals using the properties of deposit.

Materials & Methods 180 samples of surface sediments were taken from the Chahnimeh reservoir and they were transferred to lab under standard conditions. Total Zinc concentration, deposit properties and Zinc five bonds with deposit were measured. Efficiency of the ANN and Multi-Layer Perceptron (MLP) model were evaluated to estimate the Zn bonds following the measurement of parameters in the laboratory.

Findings Five links were predicted with the aid of ANNs and MLP model. Deposit properties and total concentrations of heavy metals were considered as input and each of bonds were considered as output

Conclusion Ultimately, the ANN showed good performance in the predicting the determination of coefficients or R² (0.98 to 1) and root mean square error or RMSE (0.7 to 0.01).

Keywords Artificial Neural Networks; Heavy Metals; Sediment Pollution; Chahnimeh

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1-Introduction

Sediments are one of the main components of water reservoirs in which heavy metals can exist [1, 2]. Accumulation of heavy metals in environments such as soil and sediments can be a potential danger for human health and life [3, 4].

These metals can enter food chains [5, 6] and negatively affect the ecosystem and environmental health [7-9]. Presence of heavy metals in sediments is caused by erosion of soil or stone or human activities [10-13]. Today, measurement of the total concentration of heavy metals is a poor indicative of the environmental status, because metals make various bonds with sediments. In addition, the mobility and availability of metals are different given their physiochemical forms [1, 11, 14, 15]. Sequential extraction processes suggested by Tessier *et al.* [15] in the form of 5-stage processes are widely used for the prediction of these forms. They include exchangeable, carbonate-linked, iron manganese-linked and carbonate-linked bonds together with the remaining part. Meanwhile, artificial intelligence methods have a high potential in the prediction of these forms [2, 16, 17].

Artificial neural networks (ANNs) are computer-based computational tools with a similar performance to biological processes of the human brain. These models are increasingly applied in different sciences and technologies [18-20]. Unlike most models that are based on assuming linearity between responses which deal with the prediction of variables and their normal distribution, ANNs models are able to map nonlinear connections based on variables in an ecological environment [21-23].

In a neural network, learning means the determination of optimal values of weights and other parameters such as the bias and the slope of stimulation function to have the maximum efficiency [24]. In functional estimation targets in which the neural network is in charge of establishing a connection between the groups of input and output data, the network efficiency is specified by defining the error between the real output and the network output for the set of training and test data. In the network learning, the aim is the minimization of this error by proper variation of weights and other parameters of the network. A common method widely used in this regard is called feed forward neural networks (FNN) by the training

algorithm of Levenberg–Marquardt (LM). This method has the highest efficiency compared with other neural network methods, thus it was chosen for this research. In case there are sufficient numbers of layers and neurons in the layers of these networks, they are able to estimate every nonlinear function with a desirable accuracy [25]. Perceptron networks benefit from the learning rule of post-emission error, which is a generalized algorithm of normal least squares.

So far, various researchers have successfully used ANNs for environmental studies for various purposes [26-31].

Mohammadi *et al.* [32] in the similar work developed, the neural-fuzzy model (Subtractive clustering), for the prediction of lead bonds in Chahnimeh 1, Zabol, was able to account for over 99% of lead bonds in the sediments; considering statistical criteria of root mean squares error (RMSE; 0.0337–0.0813) and determination coefficient or R^2 (0.92-0.99), this model showed good performance with regard to prediction.

2- Objective

Even though different studies have been conducted for modeling and predicting heavy metals present in sediment with the aid of different methods, there are still numerous unresolved problems in this field, including the selection of the best combination of input variables of the model for accurate prediction of Zinc bonds in sediments of water reservoirs. For instance which variables are suitable for modeling as input; and what percentage of observational data is required for model training as well. Thus, the aims of this research are to find the optimal combination of input variables in the modeling, detect the optimal number of data related to the model training, and estimating of Zn bonds using ANNs method in the surface sediments of water reservoir 1 of Chahnimeh located in the Sistan plain.

3- Materials and Methods

3- 1- Description of the study area:

Chahnimeh reservoirs in Zabol, 3 natural reservoirs are located in the south of Sistan plain and have covered an area of 50km². These reservoirs supply the water required for two cities of Zahedan and Zabol with a population of around one million people. The water of these reservoirs is provided by Hirmand River by a canal with a capacity of 1000m³/s [33].

3- 2- Sampling: In this study, 180 samples were collected from sediments in Chahnimeh 1. As the sampling was carried out using grids of 500×500-m intervals (Figure 1), it was able to provide a proper image of the studied area. The results provided the mean of the 180 measured points all samples were sealed in clean polyethylene bags and brought back to the lab to be air-dried and stored until further use.

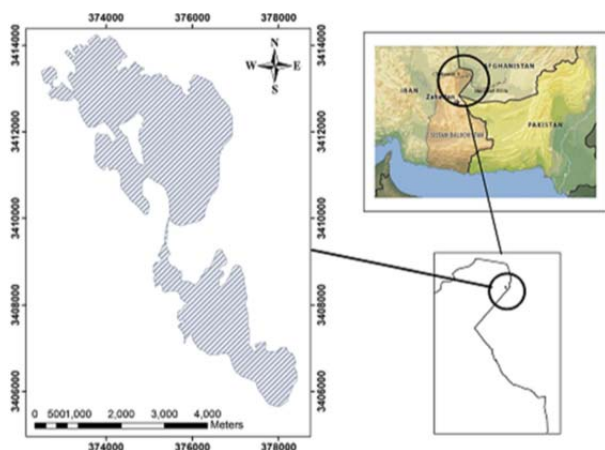


Figure 1) Map showing the geographical setting of the Chahnimeh 1 reservoir

3- 3- Experimental methodology: To measure the level of heavy metals in sediment samples, first the samples should be prepared in an atomic absorption device (Shimadzu AA-7000 spectrophotometer; Kyoto; Japan). For this purpose, the clay part of soil which is the same as the particles sieved through a 63-micron sieve was used. After digestion of samples by hydrofluoric acid (HF) 48% (Merck; Germany), H_2SO_4 95-97% (Merck; Germany), $HClO_4$ 70-72% (Merck; Germany), HCl 6 normal (Merck; Germany), HNO_3 70% (Merck; Germany), the concentration of all metals was measured by a spectrophotometer instrument (Shimadzu AA-7000 spectrophotometer; Kyoto; Japan). After measurement of the total concentration, five other experiments were conducted culminating in liberation of 5 bonds of the metal total concentration from the sediment through Tessier method [15]. Tessier method [34]. The detailed procedures employed in this process were the following:

1) Exchangeable fraction: A total of 1g of the air-dried sediment sample was extracted with 20ml of $MgCl_2$ (1mol/lm; pH=7) for 16h at room temperature (25% to 30°C) under agitation at 160rpm using a rotary bed.

2) Fraction bound to carbonates: The residue

from step 1 was extracted with 20ml of 1M NaOAc (Adjusted to a pH of 5 with HOAc) for 5h at room temperature under vigorous agitation using a rotary bed.

3) Fraction bound to hydrous Fe-Mn oxides: The residue from step 2 was extracted with 20ml of NH_2OH and 4M HCl in 25% (v/v) acetic acid in a water bath (96°C) with occasional agitation.

4) Fraction bound to organic matter and sulphides: The residue from step 3 was extracted with 3ml of 2M HNO_3 and 30% H_2O_2 (Adjusted to a pH of 2 with HNO_3) for 2h in a water bath (85°C) with occasional agitation. Subsequently, 3ml of H_2O_2 was added to the extracted solution and left for 3h at 85°C. Then, 15ml of 3.2M NH_4OAc in 20% HNO_3 was added to the solution and shaken continuously for 30min at room temperature.

5) Residual fraction: the residue from step 4 was extracted with 8 ml of aqua regia ($HCl+HNO_3$).

These five bonds included those that were exchangeable, as well as those linked to carbonate, iron, and manganese, and organic compound bonds, together with a bond with the remaining part.

3- 4- Artificial neural networks (ANNs):

Basically, an ANNs model consists of three distinct layers. The input layer is introduced to the model by input data. The weighting of all input layers is almost done in this layer. A hidden layer or layers in which data are processed, and the output layer out of which output is produced. Every layer contains one or more basic elements called a neuron or node. Each neuron has a threshold and an activity function playing a role in the training process [35, 36]. A feed forward neural network is a subgroup of network layers with no intralayer relationships, but establish a relationship from the neuron i to $i+1$ [37]. The number of input and output units is dependent on the number of input and output variables of the data [38].

3- 5- Categorization of data and Analysis:

In this research, 70% of the data were selected for training, 10% for validation, and 20% as test data for the model. In the MLP neural network, the training principle of Levenberg-Marquardt was used for training together with Tribas transfer and tangent hyperbolic functions. The number of neuron in the hidden layer was determined through trial and error. Finally,

modeling was performed by MATLAB 7.12 Software (MathWorks, Inc., Natwick, MA).

In order to compare the values of physical properties predicted by the models applied in this research with the values measured in the laboratory and to compare the accuracy of the existing models, the parameters of Coefficient of Determination (R^2) and Root Mean Square Error (RMSE) were utilized (Relation 1 and 2).

$$1) \quad R^2 = 1 - \frac{\sum_{i=1}^N (Z - \hat{Z})^2}{\sum_{i=1}^N (Z - Z^*)^2}$$

$$2) \quad RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (Z - Z^*)^2}$$

Where, $\hat{Z}$ is the estimated values at the i th point, Z^* is the mean of values predicted for the properties, Z is the values observed for the i th point, and N is the number of studied samples. Data analysis was performed by Excel.

4- Results and Discussion

The present research studies the relationship between some properties of sediments and

predicts the quintuple bonds of heavy metals with the sediment using a neural network.

The descriptive statistics for the total concentration of Zn, Zn fractions, and other sediment properties of the studied regions were calculated, as provided in Table 1. The content of the total concentration of Zn in the studied sediments had the highest dispersion (804.52) with an average of 98.6. The results obtained from the normal test indicated that the distribution of studied properties in Table 1, apart from organic carbon percentage and percentage of sand particles, follows a normal distribution. Correlation analysis (Pearson coefficient) between the total concentration of Zn and some properties measured by Zn fractions was conducted by SAS Software. In this analysis, the total Zn concentration presented the highest positive correlation (Pearson correlation coefficient) and the cation exchange capacity showed a negative correlation (-15%) with Zn fractions (Table 2).

Table 1) Some statistics of the sediments properties in the studied region

	N	Min	Max	Range	Mean	Std. Deviation	Variance	Skewness	Kurtosis
CEC	180	1.0	79.0	78.0	37.2	16.6	277.0	-0.05	-0.55
OC%	180	0.1	1.5	1.4	0.3	0.1	0.0	2.41	11.55
Sand%	180	0.4	98.7	98.3	15.1	19.5	382.4	2.09	4.00
Clay%	180	1.0	71.0	70.0	45.9	16.7	280.5	-0.73	-0.35
silt%	180	0.3	78.0	77.7	38.9	10.0	101.6	-0.30	2.65
Znf1	180	0.3	4.8	4.5	2.0	0.5	0.3	0.22	3.36
Znf2	180	1.8	6.5	4.7	3.6	0.5	0.3	0.22	3.36
Znf3	180	9.2	1.4	7.8	4.7	1.7	312.1	0.89	3.57
Znf4	180	4.4	1.3	3.1	7.7	1.0	1.1	0.22	3.36
Znf5	180	12.1	79.4	67.3	38.0	8.5	73.3	0.21	3.35
Zn total	180	30.6	243.9	213.3	98.6	28.3	804.5	0.68	3.34

Table 2) Correlation between some properties of the Chahnimeh floor sediments and Zn fractions

	Clay	Silt	Sand	O.C	CEC	Zn total
Zn F1	-0.10	0.11	0.03	0.04	-0.15	0.99
Zn F2	-0.10	0.11	0.03	0.04	-0.14	0.99
Zn F3	-0.10	0.11	0.03	0.07	-0.15	1.0**
Zn F4	-0.10	0.11	0.03	0.04	-0.15	0.99**
Zn F5	-0.10	0.11	0.03	0.04	-0.14	0.99**

The results of this study for total concentration of Zinc are consistent with the results of some other investigations [39, 40]. Also, the results for the five links for two first phases match with the results reported by Li *et al.* [41]. The maximum amount of released metal was in

bonds 3 and 5 which corresponds the results of other researchers [41-43].

In this research, in order to predict Zn fractions by Multilayer Perceptron ANN method, a general model was considered using parameters including organic carbon percentage (OC%), clay percentage (Clay%), Silt percentage (Silt%), cation exchange capacity (CEC), and total concentration of Zn. In this model, the mentioned parameters were regarded as network inputs and the measured components of Zn were considered as the target. Note that during the training process of the models, the most appropriate number of neurons in the hidden layer and the most

suitable function again in the hidden layer were chose through trial and error in order to increase the accuracy of training processes. The results of modeling are provided in Table 2. So as to train the data at the training stage of Levenberg–Marquardt (LM) algorithm was utilized. The results obtained from the structures designed in the MLP neural network architecture suggested that the most suitable functions used in the hidden layer were of Tribas and Tansig type. In the outer layer, the linear function of Purelin was used Table 3.

According to Table 3, with regard to modeling of different components of Zn using a general model (Including 5 inputs) by MLP neural network model, it can be stated that the MLP neural network, with low RMSE and high R² values, presents a very good performance in the modeling and prediction of Zn fractions. The results obtained from Table 3 also suggest that Zn fractions modeling is also possible using other properties of sediments and as evident diagram 1, in the modeling created by the MLP neural network, the predicted data are highly congruent with the experimental data. This follows that the modeling developed to estimate Zn fractions using the inputs of interest has been well able to estimate the content of Zn fractions.

In any case, with regard to the modeling and estimation of Zn fractions, the performance of the MLP neural network can be described as approvable. One thing worth mentioning here is

that artificial intelligence-based computational methods have acted very successfully in this research in terms of modeling. They have been able to consider complex and nonlinear relationships between Zn fractions and the desired inputs (Organic carbon percentage, cation exchange capacity, total Zn concentration, and the percentage of clay and silt particle concentrations).

Table 4 presents some statistics about the experimentally measured data used at training stages and test using the MLP neural network model. In general, careful investigation of these statistics indicates the validity of the modeling and estimation of Zn fractions in the studied region. As can be observed, the dispersion range (Maximum and minimum) of the data utilized at the training and test stages using the MLP neural network model is equal in most cases, and only in a few cases there is a slight difference between the averages of data used at the training and test stages.

In line with the forecast of heavy metals in sediments, predicting the concentrations of whole heavy metal with deposit characteristics was done by some researchers who have obtained successful results as the present one [44, 45].

The results of the sequential extraction indicated that each heavy metal had various risk percentages in the five phases and therefore should be taken into account for different heavy metals.

Table 3) Description of the structure of models used in Zn fractions modeling and the results obtained from Zn fractions modeling using some properties of soil

Model/ Inputs	output	MLP Structure				Train		Validation		Test	
		Neorun in hidden layer	HLTF*	OLTF*	Description	RMSE	R	RMSE	R	RMSE	R
MLP1 OC, Clay, Silt, CEC, Zn total	ZnF1	4	Tribas	Purelin	LM algorithm**	0.23	0.76	0.17	0.90	0.18	0.80
MLP2 OC, Clay, Silt, CEC, Zn total	ZnF2	4	Tansig	Purelin	LM algorithm	0.10	0.90	0.10	0.95	0.17	0.71
MLP3 OC, Clay, Silt, CEC, Zn total	ZnF3	7	Tribas	Purelin	LM algorithm	0.14	0.86	0.08	0.94	0.14	0.84
MLP4 OC, Clay, Silt, CEC, Zn total	ZnF4	6	Tansig	Purelin	LM algorithm	0.12	0.87	0.13	0.89	0.12	0.88
MLP5 OC, Clay, Silt, CEC, Zn total	ZnF5	6	Tribas	Purelin	LM algorithm	0.11	0.88	0.09	0.96	0.11	0.89

*HLTF (Hidden layer transfer function), OLTF (Output layer transfer function); **LM algorithm (Levenberg-Marquardt algorithm)

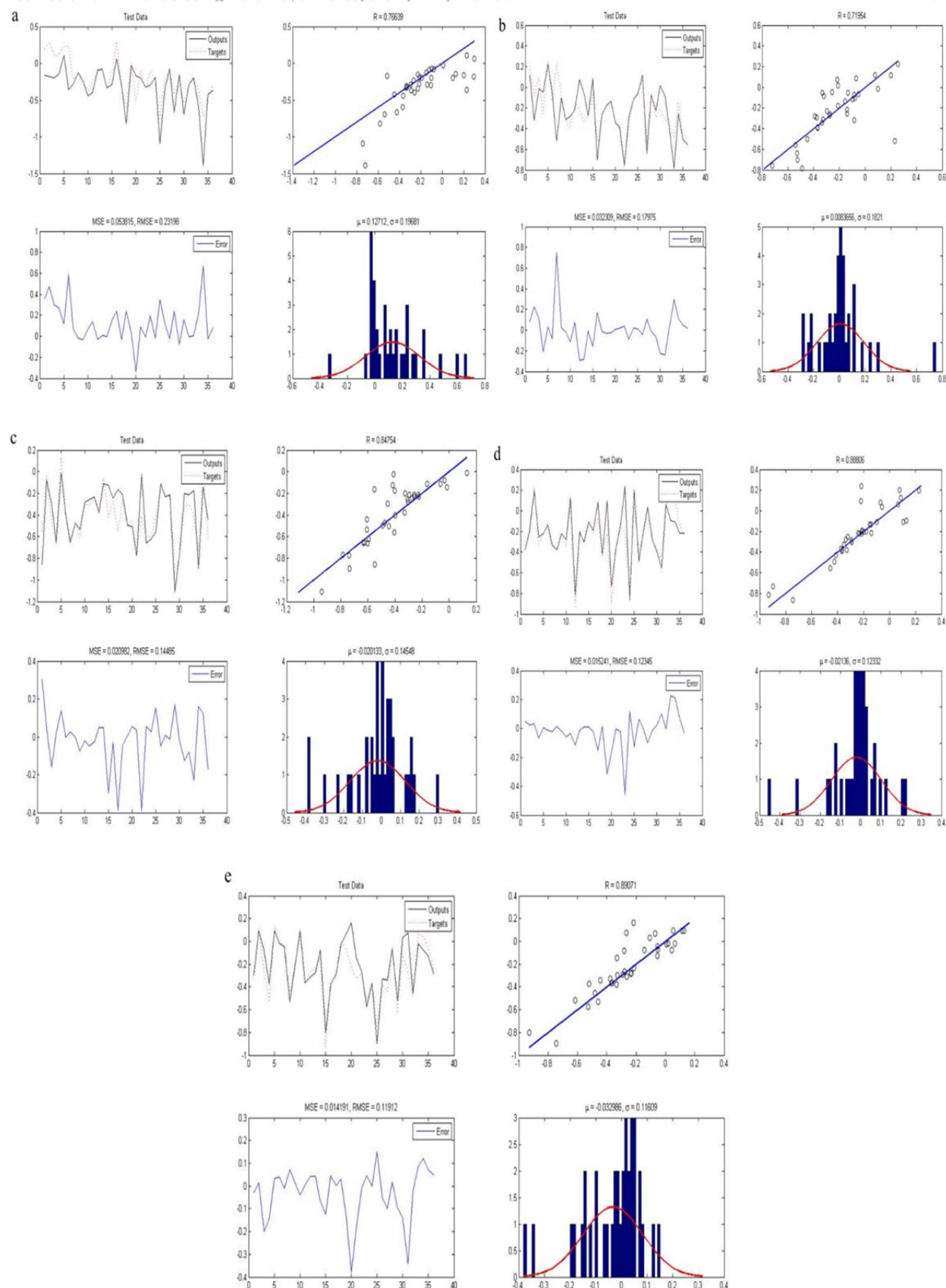


Diagram 1) The diagrams of the comparison of measured and predicted values for Zn fractions modeling using some properties of Chahnimeh floor sediments at the Test stage (a:MLP1; b:MLP2; C:MLP3; d:MLP4; e:MLP5)

Table 4) Statistics about the input data at the training and test stages, used for modeling and prediction of Zn fractions

Level/ Model/ Des.	CEC	O.C	Clay	Silt	Zn Total	Zn Fractions
MLP1						
Train						
Max	79.0	1.0	70.1	78.0	243.8	4.8
Min	1.0	0.1	1.0	0.3	30.6	0.3
Mean	36.3	0.3	45.2	38.5	97.2	2.0
Test						
Max	75.0	1.5	70.1	64.0	155.4	3.1
Min	5.0	0.1	5.0	24.0	44.3	1.0
Mean	40.3	0.3	49.1	40.9	105.4	2.2
MLP2						
Train						
Max	79.0	1.5	71.0	58.0	165.1	5.0
Min	1.0	0.1	1.0	0.3	30.6	1.8
Mean	36.5	0.3	45.9	38.1	97.9	3.6
Test						
Max	70.0	1.0	68.1	78.0	243.9	6.5
Min	2.5	0.1	6.1	6.9	60.9	2.9
Mean	38.0	0.2	44.5	41.2	102.7	3.7
MLP3						
Train						
Max	79.0	1.5	70.1	78.0	243.9	140.0
Min	1.0	0.1	1.0	0.3	30.6	9.2
Mean	38.0	0.3	46.4	38.5	99.1	47.3
Test						
Max	65.0	0.7	69.6	64.0	152.3	81.2
Min	2.5	0.1	13.0	24.9	31.2	12.5
Mean	35.7	0.3	45.9	42.3	97.0	46.0
MLP4						
Train						
Max	79.0	1.5	71.0	78.0	243.9	13.0
Min	1.0	1.0	1.0	0.3	30.6	4.4
Mean	37.1	0.3	45.3	38.5	99.1	7.7
Test						
Max	70.0	0.5	68.0	60.0	137.6	9.1
Min	5.0	0.1	6.1	6.9	62.4	6.4
Mean	37.8	0.2	47.5	38.6	93.2	7.5
MLP5						
Train						
Max	79.0	1.5	71.0	78.0	243.9	79.4
Min	1.0	0.1	1.0	0.3	31.2	12.1
Mean	37.7	0.3	46.3	39.2	98.2	37.9
Test						
Max	73.0	1.0	69.6	60.0	165.0	57.1
Min	2.5	0.1	5.0	20.0	56.0	26.2
Mean	37.9	0.3	49.6	39.8	105.0	40.1

5- Conclusion

In the present research, modeling and estimation of Zn fractions were evaluated and investigated using parameters including organic carbon percentage (OC%), clay percentage (Clay%), silt percentage (Silt%), cation exchange capacity (CEC), and the Zn total concentration by the MLP neural network

method. Overall, the results indicated that the MLP neural network has had a very desirable performance in the estimation of Zn fractions.

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