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NATURAL WATERS

Assessing the Presence of Metals in Surface Waters: a Case Study Conducted in Algeria Using a Combination of Artificial Neural Networks and Multiple Indices

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Abstract—Elevated concentrations of heavy metals in wetlands can contaminate surface water, posing hazards to human health and ecological balance. Given increasing urbanization and activities in places like Algeria, it is crucial to closely monitor and effectively control heavy metal pollution in surface water. This study proposes the use of artificial neural networks (ANN) and various indicators to comprehensively assess metal contamination in Algerian surface waters and its implications for public health. Sixteen water samples were collected for the composition analysis and source identification. Measurements indicated that several areas exceed the World Health Organization (WHO) limits for four metals. Methods such as the Heavy Metal Evaluation Index (HEI) and Heavy Metal Pollution Index (HPI) were employed to assess pollution levels. Results showed that over 99% of samples exhibited significant pollution according to HPI, with 60% showing elevated pollution levels by HEI, highlighting substantial contamination risks. Principal Component Analysis (PCA) revealed that the first two components accounted for 93.540% of total variation, with subsequent components contributing 6.459% or less. PCA 1 and PCA 2, representing 49.084 and 44.456% of variability, respectively, were identified as primary components, while PCA 3 and PCA 4 each contributed less than 5.015 and 1.444% to total variance. The study demonstrated minimal error values and R^2 values exceeding 0.5 during the testing of heavy metal models, indicating robust performance. Overall, this study underscores the prevalence of elevated metal levels in water bodies, providing comprehensive insights into heavy metal contamination in Algerian basins to assist environmental management decisions and protect public health.

Keywords: heavy metals, pollution index, artificial neural networks (ANN), contamination, Algeria

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INTRODUCTION

Water contamination is a pressing global environmental concern due to humanity's heavy reliance on surface water as a primary water source. The presence of toxic substances and other stressors significantly impacts the health of freshwater ecosystems, leading to a decline in their fundamental functions [1]. Water quality is crucial for maintaining the health and functionality of terrestrial ecosystems, encompassing various chemical, physical, and biological characteristics that vary by season and geographical location. Groundwater plays a vital role in rural areas as a primary drinking water source, contributing significantly to socio-economic development [2]. Human-induced contaminants, particularly heavy metals, pose a major threat to water sources due to their high toxicity even at minimal concentrations. Industrial activities, improper solid waste disposal, effluent discharge, urbanization, and mineral leaching are primary contributors to heavy metal pollution [3].

Studies have consistently shown elevated concentrations of heavy metals in stream sediments, largely from anthropogenic sources entering river tributaries [4]. Surface sediments act as reservoirs for metals that can be released into overlying waters, posing risks to river ecosystems [5]. Inadequate waste management infrastructure and unregulated dumping near urban areas contribute to significant levels of heavy metal contamination in rivers [6]. Lakes, particularly those with limited self-purification capacity, are particularly vulnerable to heavy metal accumulation, which can persist and bioaccumulate in organisms over time [7]. Manganese (Mn), chromium (Cr), cobalt (Co), iron (Fe), arsenic (As), nickel (Ni), and cadmium (Cd) concentrations in surface waters often exceed drinking water standards globally, posing serious threats to ecosystems and human health [8].

Algeria, with its extensive salt lakes (Sabkha or Chott), faces unique environmental challenges, especially in the southern regions where favorable weather conditions facilitate the formation of sodium chloride salt crystals through solar evaporation [9]. Rapid population growth and industrialization have led to significant environmental degradation in Algeria's urban and industrial centers like Arzew, Annaba, Skikda, and Algiers, where untreated waste disposal and maritime activities further exacerbate water quality issues. Despite increasing scrutiny, limited research has been conducted on metal concentrations in Algerian surface sediments, with few studies addressing ecological risks and biological impacts [10]. For instance, previous studies in Ghazaouet focused on localized contamination from zinc electrolysis and tailings leaching, offering insights into specific areas rather than a comprehensive overview of Algeria [11].

This study aims at enhancing the understanding of pollution origins and impacts on Algeria's ecosystems, focusing on 16 water bodies across different regions. Selected for their ecological significance, the study seeks to identify sources of pollution, particularly heavy metals, and quantify levels of significant pollutants.

MATERIEL AND METHODS

Study Area

Algeria is located in the North Africa, spanning an expanse of 2.381.741 km^2. It stretches along the Mediterranean Sea from east to west, covering approximately 1,200 km, and from north to south, spanning about 2.000 km. Over the past 30 years, Algeria's population has surged by 95%, rising from 22 million in 1985 to 44.6 million in 2023, as reported by the National Statistical Bureau in 2023. This population surge exacerbates the strain on the ecosystem, particularly in regions near major urban centers such as Algiers and Oran, and in proximity to industrial zones and ports like Arzew Salt, Ghazaouet, Bejaia, and Skikda. These pollution sources on the Algerian coast include household waste, discharges from the chemical and petrochemical industries, and emissions from thermal power plants.

The primary aim of our study is to conduct a thorough investigation into heavy metal pollution in wetland areas within Algeria. Specifically, our focus will be on various water features within Algeria's wetland complex (Fig. 1), which includes significant sites such as Macta Marsh, Great Sebkha, Telamine Lake, Arzew Salt, Boughezoul Dam, Chot Ech Chergue, and Dayet El Ferd. These sites are designated under the Ramsar Convention on Wetlands, underscoring their ecological importance. Additionally, we will give particular emphasis to additional sites such as Beni Bahdel Dam, Bougara Dam, Bouhanifia Dam, Chorfa Dam, Cheliff Dam, Dahmoni Dam, Gargar Dam, Karrada Dam, and Sarno Dam (Table 1).

The study area comprises multiple watersheds that share consistent environmental conditions. The water quality across this region is uniformly described as saline, which is indicative of the prevalent geological and hydrological influences shaping the area's unique ecosystem dynamics. Furthermore, the study area falls within a Semi-arid climate classification, highlighting its susceptibility to water scarcity and seasonal variability. These climatic conditions have a profound impact on hydrological patterns and ecological processes within the watersheds, influencing the distribution and concentration of heavy metals in the wetland environments.

The wet areas selected for this study are chosen based on their environmental and economic significance, as classified by the Ramsar Convention, which designates them as areas deserving preservation and sustainable use. The investigation aims to offer insights into the risks posed by heavy metal pollution to these unique wetland features and to contribute to their long-term conservation and sustainable management.

Sample Collection and Analysis

From February to March 2023, surface water samples were collected for an environmental quality study. Each sample consisted of 1.5 L of water collected in specially cleaned plastic bottles and rinsed thoroughly with double-distilled water to prevent contamination. The samples were carefully labelled to ensure quick and accurate identification of their origin and were stored in certified ice boxes at 4°C to maintain stability during transport and storage. To prepare the samples for heavy metal analysis, they were acidified using 0.5% nitric acid (HNO_3) to maintain a pH value below 2.0. According to paper [12], this acidification process prevents the precipitation of metals and minimizes the adsorption on container surfaces.

Subsequently, the samples were transported to the laboratory at the Scientific and Technical Center for Physical and Chemical Research (CRAPC) in Algeria. Various parameters such as pH, temperature,

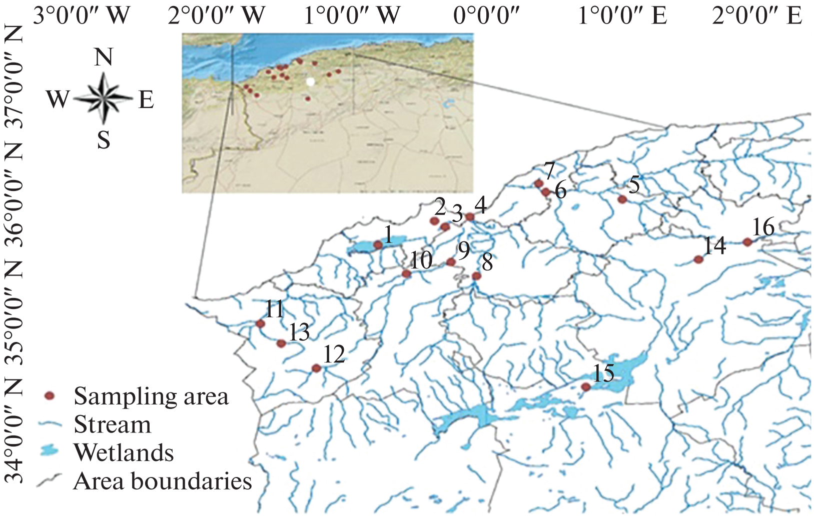

Fig. 1. Cartographic Display of Water Networks in Algeria, Basin Stations, Sampling Stations, and Supply Water.

S, EC, and TDS were measured on-site using the multifunctional HI 9811-5 device. In addition, concentrated samples underwent analysis to determine the concentrations of heavy metals. AAS was employed for accurate quantification of these elements.

Metal Assessment Index

To determine the degree of metal pollution in various aquatic systems, HEI is usually used and estimated as [13]:

$$\mathrm{HEI} = \sum_{i=1}^{n} \frac{H_c}{H_{\mathrm{Mac}}}, \tag{1}$$

Table 1. Characteristic Sampling Stations: Codes and GPS Coordinates

Watershed	Watershed, ha	GPS coordinates
Great Sebkha	56.870	35°31′29″ N 00°47′12″ W
Telamine Lake	5.778	35°44′09″ N 00°22′57″ W
Arzew saline	44.500	35°41′25″ N 00°19′22″ W
Macta Marsh	358.280	35°38′52 N 00°06′16″ W
Gargar dam	50.000	35°55′52″ N 0°59′15″ E
Cheliff Dam	65.000	35°59′00″ N 0°24′47″ W
Karrada dam	34.52	36°05′1162″ N 00°38′5519″ E
Bouhanifia dam	70.210	35°16′39″ N 0°3′36″ W
Chorfa dam	21.250	35°25′55″ N 0°14′43″ W
Sarno dam	175.450	35°29′94′99″ N 00°59′0768″ W
Boughrara dam	3.323	34°88′3854″ N 01°67′6169″ W
Dayet El Ferd	54.630	34°29′55″ N 01°14′23″ W
Beni Bahdel dam	39.520	34°42′42.70″ N 1°30′13.24″ W
Dahmoni dam	855.500	35°41′67″ N 14°76′29″ E
Chott Ech Chergui	11.320	34°16′09″ N 00°33′25″ E
Bougara dam	11.320	36°32′26″ N 3°5′7″ E

where H_{Mac} and H_{c} are the assigned values for the i-th parameter and the maximum permissible concentration, respectively.

Heavy Metal Pollution Index

Various statistical methods were employed to clarify the data for better comprehension. HPI is a grading system that illustrates the cumulative impact of multiple metals on water resource health. This rating ranges from 0 to 1 indicating the relative individual quality conditions, and is defined as the inverse of the recommended standard (Si) for each parameter. Overall, the water quality can be assessed by computing its quality index. This study employed WHO standards for permissible values in drinking water [14]. The HPI was computed using the following equation:

$$\text{HPI} = \frac{\sum_{i=1}^{n} W_i Q_i}{W_i}, \tag{2}$$

where W_i is the unit weight of the i-th parameter, Q_i is the subindex of the i-th parameter and n is the number of parameters considered. The value of Q_i is determined by the following equation:

$$Q_i = \sum_{i=1}^{n} \frac{(M_i - I_i)}{(S_i - I_i)} \times 100, \tag{3}$$

where M_i is the monitored value of the heavy metal of the i-th parameter, I_i is the ideal value of the i-th parameter, and S_i is the standard value of the i-th parameter.

Statistical Analysis

This study utilized the software XLSTAT 2024 for statistical analysis. The relationship between different elements was assessed through the Pearson Correlation Analysis. Additionally, HCA was employed to group sample points based on their metal concentrations. Variables were divided into clusters using variable distances, employing the intergroup linkage method and the square Euclidean distance, known for its stability in systematic clustering analysis. PCA was also conducted to determine how much variance each variable explains after the dimension reduction. PCA explored the relationship between elements by extracting potential factors and simulating similarity in the distribution of heavy metal sources [15].

Artificial Neural Network

ANN is a type of nerve examination that works using input data. ANN is often used in scientific research thanks to its ability to perform classification and regression on a complex non-linear dataset [16]. This structure consists of three important parts: the input layer, the hidden layer, and the output layer. Many nodes have variable weights and biases that they can learn in hidden layers. Hidden layers are applied to capture the information contained in anomalies. The activation function transfers the flow of data from each node to the next layer, enhancing the linearity of the factor. During this process, weights and biases change to mitigate differences between expectations, measurements, and potential predictions. This method applies in environmental research because it provides predictions for a complex dataset [17].

Using ANN methodologies, this study analyzed the data for water samples. The complete dataset used for training and testing was divided into two parts: 70% for training the neural network and 30% for validation and testing. The training involved presenting full pattern pairs and adjusting connection weights using an iteration-based method in MATLAB Neural Network Toolbox. After training, the network was tested with data to verify the training efficiency. In ANN, this step is called "testing" because it ensures that the system has learned from specific patterns of application instead of memorizing a particular dataset. It is also known as a generalization set, which is used to validate the trained network performance on unseen data. If we can generalize outputs from test datasets, then we can predict new data correctly. To avoid an overly complex model, the number of hidden layers was reduced to one, and the number of hidden neurons was set between 1 and 30. An early-stopping approach was also used to prevent overfitting, where training is halted at the point of minimal error (Fig. 2).

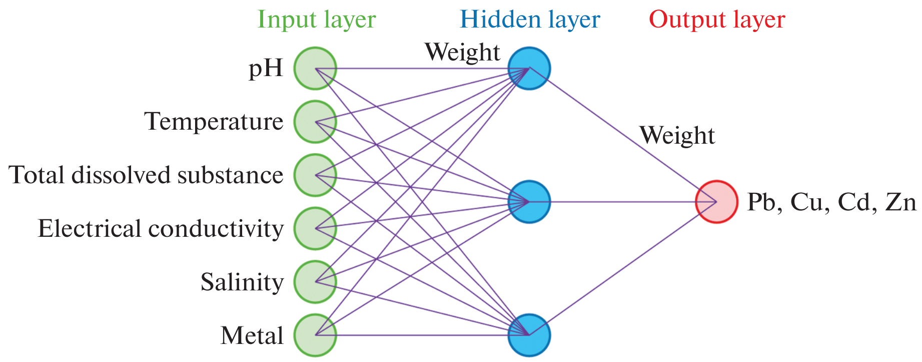

Fig. 2. ANN architecture.

RESULTS AND DISCUSSION

Heavy Metal Contamination Indices (HPI, HEI)

The pollution study assesses heavy metal levels in surface water samples from wetlands using two primary indicators: HEI and HPI. According to reference [18], HEI categorizes pollution levels as follows: HEI < 10 indicates low pollution, HEI = 10–20 indicates moderate pollution, and HEI > 20 indicates high pollution. Reference [19] classifies HPI based on concentrations of lead, mercury, cadmium, nickel, chromium, and zinc in surface water: HPI < 50 indicates minimal pollution, HPI = 50–100 indicates moderate pollution, and HPI > 100 indicates elevated pollution. HEI reflects biological responses to heavy metal contamination.

The findings reveal significant disparities in HPI and HEI levels across different monitoring locations. Many stations exhibit elevated HEI values, indicating moderate to high levels of heavy metal contamination. Stations such as Garages, Bauhinia, and Chott show particularly high HEI values, indicating significant heavy metal pollution. While HPI readings vary considerably, most stations display high to moderate HPI values, suggesting substantial heavy metal contamination. The study underscores widespread heavy metal pollution in the investigated areas, posing risks to both the environment and public health. This contamination can be attributed to factors such as industrial and agricultural activities, as well as residential and industrial waste (Table 2).

Exploring the Association between Heavy Metals, Physical-chemical Interactions, and HPI

Statistical analyses were employed to assess the presence of toxins in water samples and to investigate factors influencing the movement and distribution of metal pollutants. Metal coordination can indicate sources and pathways to particulate media [20]. The Pearson correlation coefficient (r) was used to evaluate relationships between heavy metals and water quality indicators, as well as to identify potential contaminant sources. Strong positive correlations were observed between Cu and Cd ($r = 0.848$), Cd and Zn ($r = 0.736$), and Cu and Zn ($r = 0.456$), enhancing our understanding of contaminant transport and distribution in aquatic systems. These findings underscore the significant influence of these factors on the dispersion rates and environmental impacts of toxic metals.

Furthermore, significant associations were found between physical and chemical parameters: EC and TDS ($r = 0.617$), EC and S ($r = 1$), and TDS and S ($r = 0.617$). While some variables showed positive correlations, others lacked significant associations at all levels of significance. The observed relationships between concentrations and positive associations may stem from anthropogenic pollution sources such as sewage discharge, runoff, and waste from activities like vehicle washing and industrial operations, directly entering the water system. This likely explains the notably higher concentration of Pb, which was 120 times greater than typical background levels, primarily due to mining activities [21].

However, various other sources of water pollution have likely contributed to the elevated concentrations of heavy metals observed in our study area, as detailed in Table 3. The lack of a substantial correlation between heavy metals and acidity may be attributed to differing effects of acidity levels on the chemical behavior of heavy metals in water. Moreover, the capture of heavy metals in water is influenced by complex factors, including interactions with organic matter that form soluble or insoluble compounds, affecting their mobility and availability. While several negative correlations were observed, their statistical significance was limited by weak associations.

Table 2. Evaluation of pollution indicators

Watersheds	Mean HPI	HEI
Great Sebkha	109.6506	1.937
Telamine Lake	101.8695	9.447
Macta Marsh	106.7901	4.081
Arzew saline	100.9385	40.479
Sarno dam	126.7755	5.812
Beni Bahdel dam	197.3031	5.529
Boughrara dam	266.2346	50.141
Bougara dam	132.4949	9.314
Chorfa dam	149.3703	15.198
Karrada dam	150.5508	30.131
Gargar dam	415.6029	957.942
Dahmoni dam	126.842	8.512
Chott Ech Chergui	120.5877	197.478
Bouhanifia dam	197.2699	297.818
Cheliff Dam	135.0886	10.865
Dayet El Ferd	89.66176	121.518

Table 3. Correlation analysis among heavy metals and EC, TDS, pH, S, and T in the water body

Variables	T, °C	pH	EC, s/cm	TDS	S, mg/L	Pb	Cu	Cd	Zn	HPI
T, °C	**1**									
pH	0.126	**1**								
EC, s/cm	0.028	–0.270	**1**							
TDS	0.042	0.024	0.617	**1**						
S, mg/L	0.028	–0.270	1.000	**0.617**	**1**					
Pb	–0.076	–0.284	–0.352	–0.372	–0.352	**1**				
Cu	–0.250	0.047	–0.118	–0.143	–0.118	–0.253	**1**			
Cd	–0.097	0.031	–0.237	–0.119	–0.237	–0.279	**0.848**	**1**		
Zn	0.163	–0.012	0.054	0.082	0.054	–0.318	0.456	**0.736**	**1**	
HPI	–0.066	–0.279	–0.324	–0.353	–0.324	**0.997**	–0.314	–0.351	–0.368	**1**

Principal Component Analysis

The PCA is a robust statistical method used for data analysis and dimensionality reduction, particularly valuable for understanding the distribution of heavy metals in polluted areas. Applying PCA with Varimax rotation identifies four factors above the "elbow point", where values exceed 1 (Table 4), demonstrating its effectiveness in reducing the dataset size [22]. Eigenvalues indicate that the first two components capture approximately 93.540% of the variance, while subsequent components contribute 6.459% or less to the overall variation. Specifically, PCA 1 and PCA 2 account for about 49.084 and 44.456% of the total variation, respectively (refer to Fig. 3). In contrast, PCA 3 and PCA 4 contribute proportionally below 5.015 and 1.444% of the variation, respectively.

Absolute scores categorize trace elements, with values above 0.75 classified as "strong", between 0.75 and 0.50 as "medium", and below 0.50 as "weak". Analysis reveals that Factor 1 exhibits weak positive loads of Cd, while Factor 2 shows low positive loads of Zn elements, and Factor 3 displays low negative loads of Pb and Cu elements. Both natural processes and human activities, including geological processes and emissions from industrial sources, influence these positive ion load indicators. PCA effectively identifies major contributors as sources of human and natural contamination at the study sites [23].

Table 4. The Eigenvalues of the Principal component matrix

№	Eigen value	Percentage of variance, %	Cumulative, %
1	1.963	49.084	49.084
2	1.778	44.456	93.540
3	0.201	5.015	98.556
4	0.058	1.444	100

Wastewater discharge emerges as the primary cause of water pollution, underscoring the importance of understanding the distribution and chemical interactions of heavy metals in aquatic environments [24]. Sampling sites located in the middle and downstream areas of the study show higher concentrations of parameters, likely due to intense industrial and commercial activities in those regions.

Hierarchical cluster analysis categorizes the 16 sites into three significant groups, illustrating spatial variability across Algeria (Fig. 4). The dendrogram visually represents these groupings: Group 1 includes locations such as Great Sabkha and Arzew Saline, Group 2 encompasses ten sites including Telamine Lake, Macta Marsh, and various dams, while Group 3 comprises sites like Karrada Dam, Sarno Dam, and others. These groups reflect varying levels of pollution—low, medium, and high—based on similar characteristics and natural background sources utilized in the clustering method.

Artificial Neural Network Modeling for Predicting Heavy Metal

ANN modelling was employed to predict concentrations of Cu, Cd, Zn, and Pb ions through multiple training and testing cycles using four distinct models. The integration of EC values significantly improved model performance in estimating these metals, with pH and temperature identified as crucial input variables [25]. Models with the highest specificity coefficient and lowest mean square error were selected for each metal during training.

Using MATLAB R2023b, linear analysis was conducted on input variables and their corresponding outputs. Fig. 5 illustrates the results for Pb, Cd, Cu, and Zn, while Fig. 6 displays RMS values used to assess network performance. Mean squared error was utilized to quantify the average squared difference between observed and predicted values, serving as a critical performance metric. The training of the ANN

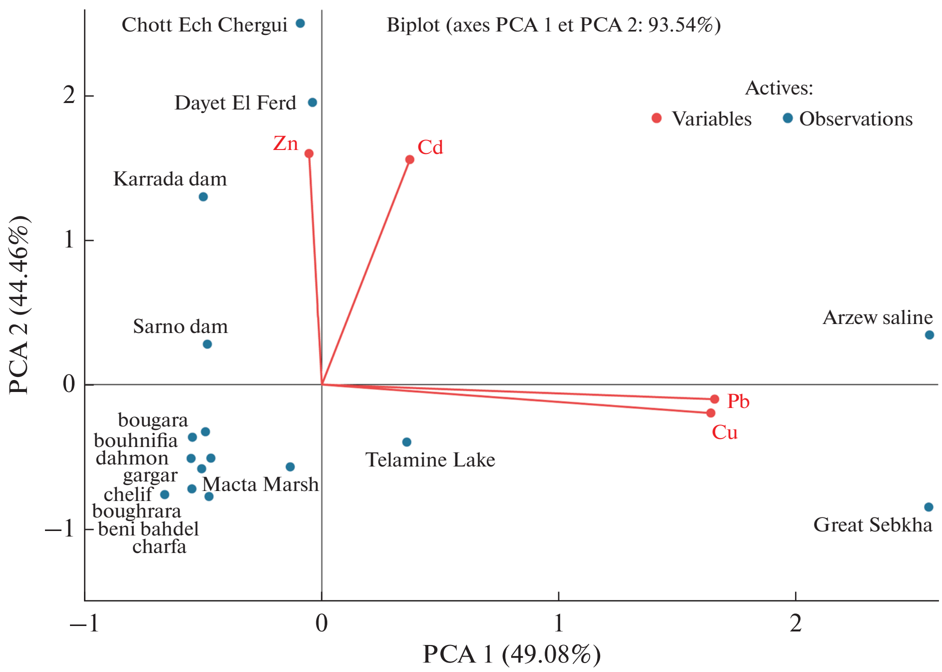

Fig. 3. Biplot visualization of PCA of heavy metal distribution across various sampling locations.

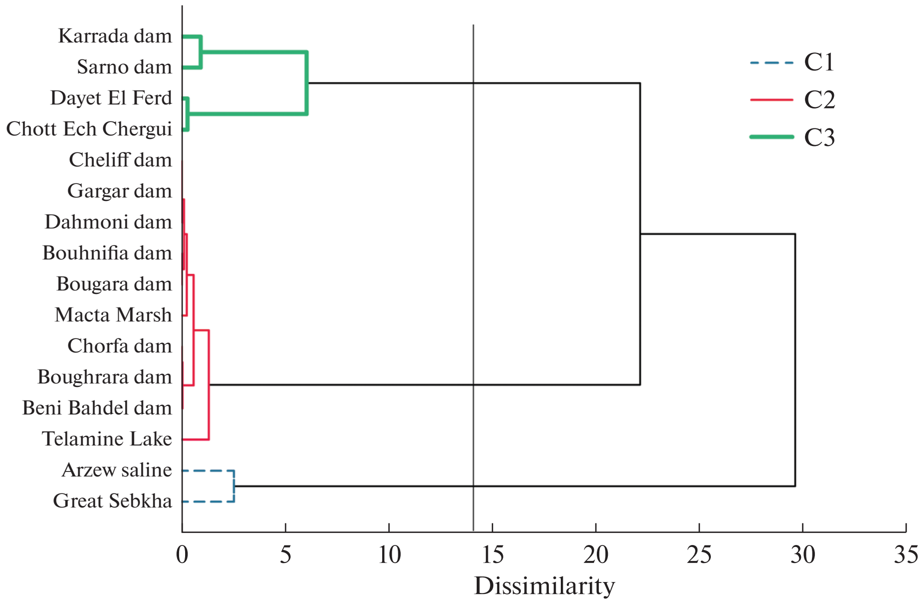

Fig. 4. Cluster analysis of sixteen sampling locations in Algeria using CAH.

models involved multiple epochs, where an epoch is defined as one complete pass through the entire training dataset. Tests and validation evaluations confirmed excellent performance, demonstrating the ability of network architecture to accurately replicate experimental data.

Table 5 presents average RMS values obtained during the development, validation, and testing phases of the proposed ANN models for Pb, Cu, Cd, and Zn. All models achieved R^2 values exceeding 0.5 during testing, indicating low error values. While the reliability varies by model type, overall, the prediction of heavy metal content using variables such as temperature, pH, EC, TDS, and S is notably effective. These ANN models, utilizing easily measurable parameters, enable rapid detection of toxic heavy metals in water bodies.

Evaluation across different heavy metals and models revealed varying degrees of performance, allowing for ranking of models from best to least suited for specific pollutants. As environmental concerns, particularly in water resources management and quality control, continue to grow, the application of ANN technology proves increasingly crucial for efficient pollution monitoring and control strategies.

CONCLUSIONS

The primary objective of this study was to assess the quality of surface water across 16 basins in Algeria, focusing particularly on heavy metal contamination. Analysis of samples collected from these surface waters revealed elevated levels of metals, indicating significant concentrations in these regions. Lead (Pb) and cadmium (Cd) emerged as the primary metals posing the greatest risk to water quality. Pollution indices indicated that a majority of the surface water samples exceeded critical pollution thresholds, highlighting significant contamination risks in these water bodies. Specifically, over 99% of the samples showed significant pollution levels according to the HPI index, while 60% exhibited high pollution levels based on the HEI, underscoring the urgent need for targeted interventions.

Table 5. Performance of the multilayer of the ANN models

Inputs	Number of hidden layer nodes	Transfer function (hidden)	Output	Transfer function output	RMS errors		
					training	validation	testing
T, pH, EC, TDS, S	3	Logsig	Pb	Tansig	0.7111	0.7596	0.0843
		Logsig	Cd	Tansig	0.0701	0.0741	0.0777
		Logsig	Cu	Tansig	0.0036	0.0017	0.0216
		Logsig	Zn	Tansig	0.0655	0.0656	0.0788

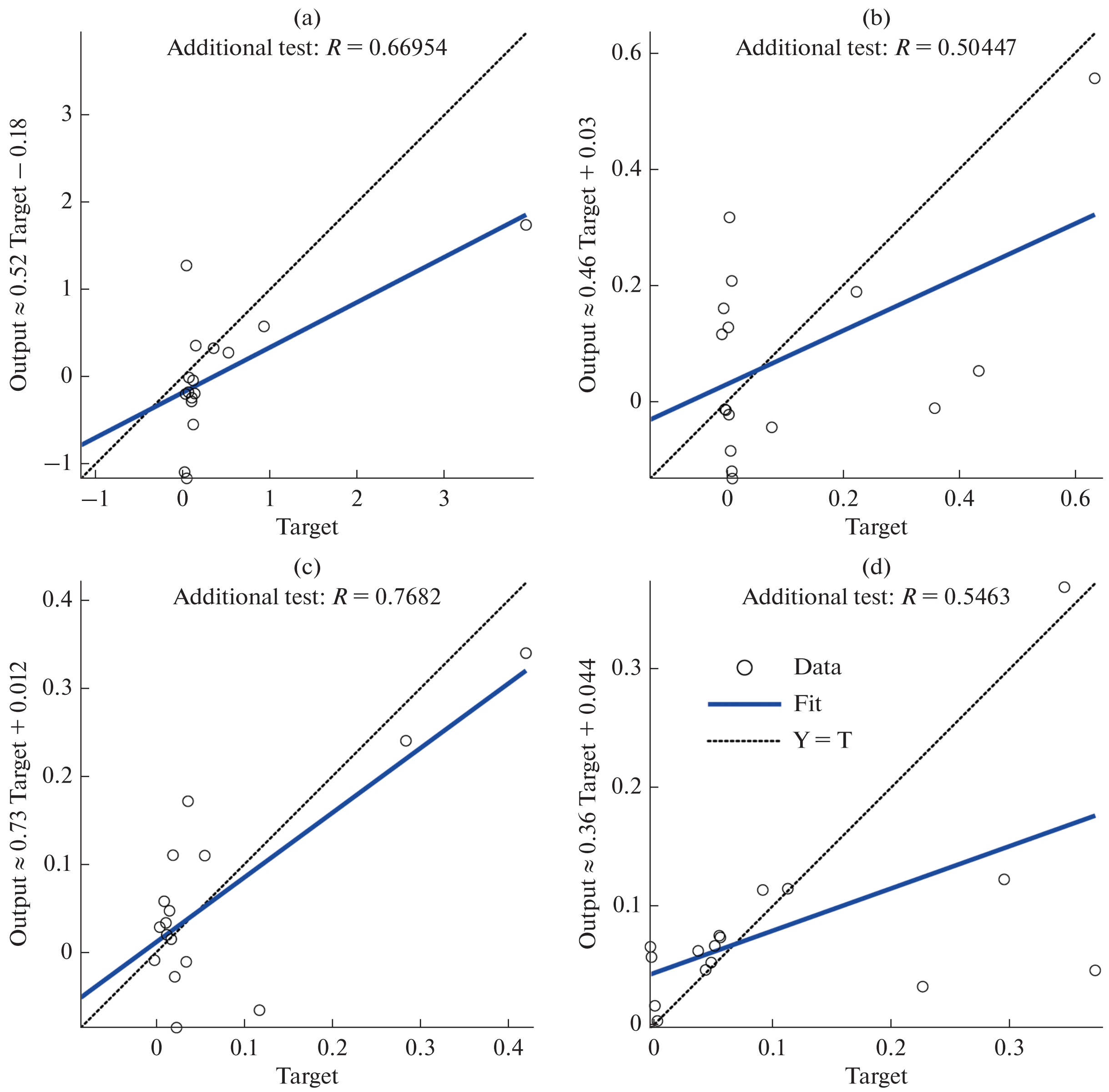

Fig. 5. Network regression for Pb (a), Cd (b), Cu (c), and Zn (d).

Through statistical analysis, The Pearson correlation, and PCA, significant sources of pollution were identified. Lead (Pb), zinc (Zn), copper (Cu), and cadmium (Cd) were predominantly linked to the mining sector with limited correlation to natural sources. Lead and cadmium were associated with emissions from chemical factories, while zinc and copper sources included fertilizers, geological formations, and human activities.

The study highlighted neural networks as an effective and economical method for predicting heavy metal concentrations, offering potential savings in time and resources for water pollution monitoring. Identifying optimal models for regulating pollution standards was a crucial outcome, suggesting the need for similar studies across different water resource categories to effectively manage and simulate pollution scenarios.

Overall, this study aims to provide valuable insights for government agencies responsible for environmental protection, particularly concerning the impact of mining activities on water resources.

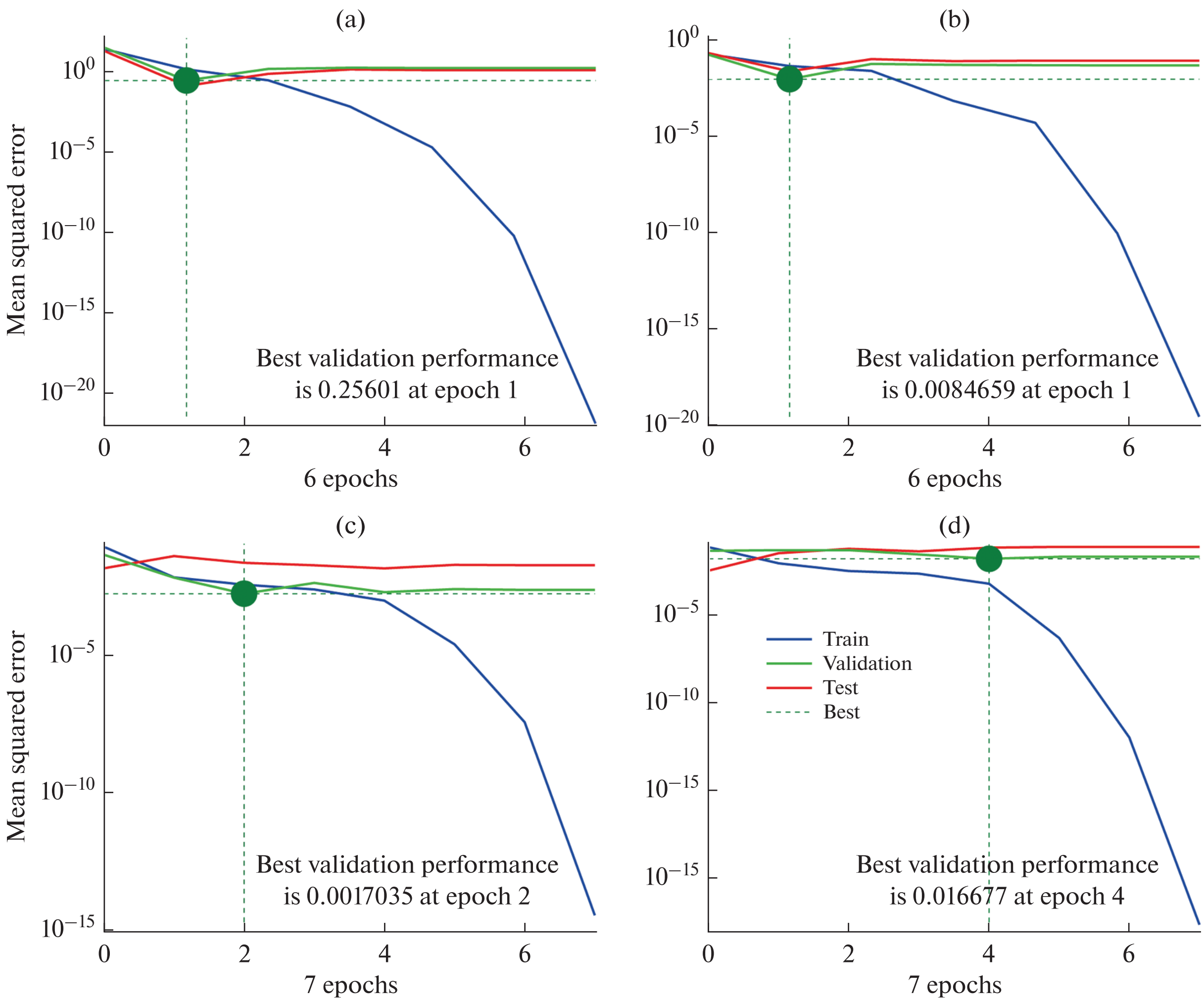

Fig. 6. RMS for Pb (a), Cd (b), Cu (c) and Zn (d).

ABBREVIATIONS AND NOTATION

S	Salinity
EC	Electrical conductivity
TDS	Total dissolved solids
AAS	Atomic absorption spectroscopy
HEI	Heavy metal evaluation index
HPI	Heavy metal pollution index
WHO	World Health Organization
HCA	Hierarchical clustering analysis
PCA	Principal component analysis
ANN	Artificial neural network
CAH	Hierarchical agglomerative clustering
RMS	Root mean square

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CONFLICT OF INTEREST

The author of this work declares that he has no conflicts of interest.

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