Modelling Nitrate Concentration of Groundwater Using Adaptive Neural-Based Fuzzy Inference System

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Abstract: High nitrate concentration in groundwater is a major problem in agricultural areas in Iran. Nitrate pollution in groundwater of the particular regions in Isfahan province of Iran has been investigated. The objective of this study was to evaluate the performance of Adaptive Neural-Based Fuzzy Inference System (ANFIS) for estimating the nitrate concentration. In this research, 175 observation wells were selected and nitrate, potassium, magnesium, sodium, chloride, bicarbonate, sulphate, calcium and hardness were determined in groundwater samples for five consecutive months. Electrical conductivity (EC) and pH were also measured and the sodium absorption ratio (SAR) was calculated. The five-month average of bicarbonate, hardness, EC, calcium and magnesium are taken as the input data and the nitrate concentration as the output data. Based on the obtained structures, four ANFIS models were tested against the measured nitrate concentrations to assess the accuracy of each model. The results showed that ANFIS1 was the most accurate (RMSE = 1.17 and R^2 = 0.93) and ANFIS4 was the worst (RMSE = 2.94 and R^2 = 0.68) for estimating the nitrate concentration. In ranking the models, ANFIS2 and ANFIS3 ranked the second and third, respectively. The results showed that all ANFIS models underestimated the nitrate concentration. In general, the ANFIS1 model is recommendable for prediction of nitrate level in groundwater of the studied region.

Keywords: Adaptive Neural-Based Fuzzy Inference System; nitrate pollution; water quality parameters

Nowadays, nitrate pollution of groundwater is an important environmental and agricultural problem in Iran. Due to the occurrence of droughts in recent years, overexploitation of groundwater for agriculture, urban and rural water supply has become an important issue in water resources management of this water-scarce region. Nitrates, being extremely soluble in water, move easily through the soil and into the groundwater (Ramasamy *et al.* 2003). Leaching of excessive amounts of nitrates has some adverse effects on infants and susceptible adults. It causes blue-baby syndrome or methaemoglobinaemia, which can lead to brain damage and sometimes to death (Ramasamy *et al.* 2003; Mahvi *et al.* 2005). The maximum permissible level for nitrates in public drinking water is established by USEPA as 45 mg/l NO_3 or 10 mg/l N-NO_3 (USEPA 2000).

Isfahan province, located in central Iran, is undergoing great land use changes due to the population growth and accompanying industrial, commercial and agricultural developments. These activities produce multiple sources of contaminants such as manure and chemical fertilizers, landfills, accidental spills and domestic or industrial wastewater (Mahvi *et al.* 2005; Almasri 2007). Among these sources, agriculture-related activities are well-known non-point source pollution. Agricultural activities may deteriorate the groundwater quality in small to large watersheds, especially due to excessive use of fertilizers and various pesticides (Almasri & Kaluarachchi 2004, 2005). Variation in groundwater quality is a function of physical and chemical parameters that are greatly influenced by geological formations and anthropogenic activities as well (Subramani *et al.* 2005). Because of the development of farmlands and overapplication of chemical fertilizers, particularly nitrogen fertilizers, nitrate has become one of the main sources of soil pollution. Therefore, it is necessary to investigate nitrate pollution of groundwater.

Artificial neural networks have been used to predict the pesticide and nitrate contamination in rural private wells (Ray & Klindworth 2000). Depth to aquifer from the soil surface, well depth and distance to cropland were used as input parameters and concentration of pesticides or nitrates was the output. A set of neural networks was also used to predict soil water content at a given depth as a function of soil temperature and soil type and was compared with a multiple regression model (Altendorf *et al.* 1999). Neural networks were generally able to predict the soil water content over time but the regression model did not perform well to follow the trend of data over time. The probabilistic, statistical, and stochastic approaches require large amounts of data for modelling purposes and therefore they are not practical in local studies. It is therefore necessary to adopt a better approach to nitrate modelling.

Terzi *et al.* (2006) investigated the prediction of pan evaporation using the adaptive neural-based fuzzy inference system. Daily evaporation, solar radiation, air and water temperatures and relative humidity measurements were used to develop the ANFIS method. The daily evaporation estimations by Penman method were used as output data for the verification of the ANFIS approach. The results from the ANFIS model had a coefficient of determination of 0.98 when compared with the Penman method results and a low average performance error of 4.6% (less than the practically acceptable limit of 10%). Keskin *et al.* (2009) compared ANFIS and fuzzy sets for their applicability to estimate evaporation from meteorological data, including air and water temperatures, solar radiation, and air pressure. The calculated evaporation values were compared with measured daily pan evaporation values. The results showed that ANFIS modelled the evaporation process successfully.

Permanent monitoring of water resources requires simple but effective nitrate concentration estimation procedures, especially from measurable groundwater data. Unfortunately, such approaches are rather scarce in the literature. Adaptive neural-based fuzzy inference system (ANFIS) was used in this paper.

In this research, the collected data on groundwater samples were used to: (1) estimate nitrate concentration in an arid region (Isfahan province, Iran) and (2) investigate the effect of some parameters on nitrate concentration, for controlling the pollution of groundwater using ANFIS.

MATERIAL AND METHODS

Study region and data

Isfahan province is located at 30°43' to 34°27'N latitude and 49°6' to 55°31'E longitude. Isfahan has arid and semiarid climates, mostly characterized by low rainfall and high potential evapotranspiration. The main river of the province, Zayandehrud, runs for some 350 km roughly west-east from the Zagros Mountains to the Gavkhuni swamp. The average rainfall of Isfahan is about 120 mm, which falls mostly in November to April. Severe droughts are recognized as a feature of Isfahan climate.

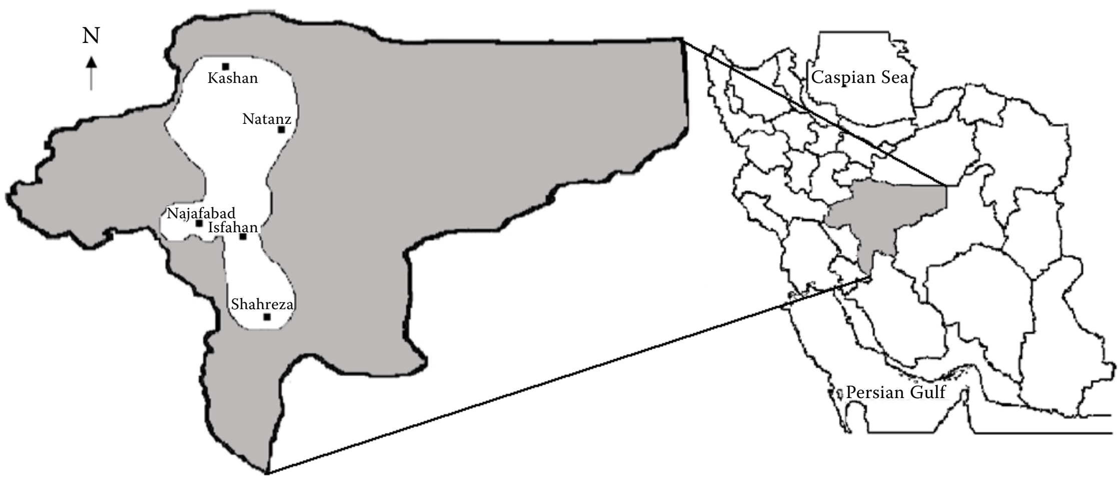


Figure 1. Location map of the region under the study

In 2009–2010, the province suffered severe dryness and this lack of rainfall resulted in extensive damage. Major crops grown in Isfahan are wheat, barley and lucerne. Isfahan also yields considerable quantities of maize, cotton, rice, soybean, grape, various nuts and vegetables. Fertilizers are applied throughout agricultural regions of Isfahan to enhance crop production. Most of the agricultural land in Isfahan is cropland and pasture. However, livestock also contribute substantially to the agricultural industry of the province. Groundwater supplies the main water consumption, with the remainder coming from numerous surface water reservoirs.

The region under investigation is a part of Isfahan province, located between northern latitude of 31°54'21'' to 34°05'31'' and eastern longitude of 51°05'30'' to 52°38'31''. This area (Figure 1) includes the cities and suburbs of Najafabad, Shahreza, Natanz, Kashan, north of the city of Isfahan and the vicinities of Zayandehrud River (Jafari Malekabadi 2002).

In this research, 175 observation wells were selected and the concentration of nitrate (NO_3^-), potassium (K^+), magnesium (Mg^{2+}), sodium (Na^+), chloride (Cl^-), bicarbonate (HCO_3^-), sulphate (SO_4^{2-}), calcium (Ca^{2+}), hardness (TH), electrical conductivity (EC) and pH were determined in a laboratory from the taken water samples. Sodium absorption ratio (SAR) was calculated from these measurements.

Selected sampling points

In this research, after delineating the study region, some representative wells were selected. Due to the extent of the study area, these wells were selected from agricultural, industrial and urban sectors. The Global Positioning System (GPS) device was used to determine the geographical locations of the wells.

Distribution of sampling points

The 175 observation wells were selected from agricultural, industrial and urban sectors of Isfahan, cities of Najafabad, Shahreza, Natanz, and Kashan and the neighbourhood of Zayandehrud River. Their distribution in different parts is as follows: 29 wells in Najafabad, 13 wells in Shahreza, 33 wells in Natanz and Kashan, and 100 wells in the neighbourhood of Zayandehrud River.

Sampling interval

Monthly water samples were taken and analysed for different chemical properties. The groundwater samples from the wells were collected in five stages, with one-month interval (January 2000 to May 2001).

Measurement techniques

In each well, the samples were taken after a pumping period of 10 min at least. The water samples were poured in clean plastic containers. They were stored in the refrigerator until they were analysed. The measurement of different chemical parameters was performed as follows: pH with a pH-meter (Model 620, Metrohm, AG Herisau, Switzerland), electrical conductivity with an EC-meter (Model 644, Metrohm, AG Herisau, Switzerland), HCO_3^- by titration with sulphuric acid and methyl orange, nitrate with an ion selective electrode (Model 3310, Jenway, Essex, UK), Na^+ and K^+ with a flame photometer (Model 410, Corning, Essex, UK), Mg^{2+}, Ca^{2+} and Cl^- by titration, and SO_4^{2-} with a spectrophotometer.

Statistical analysis

SPSS software was used to find the correlation between nitrate concentration and other chemical properties.

Adaptive Neural-Based Fuzzy Inference System

Artificial Neural Network (ANN) has been accepted as a potentially useful tool for modelling complex, non-linear systems and is widely used for prediction purposes. But, ANN is a black-box method and its inner rules are not easily understandable. The Fuzzy Inference System (FIS) involves uncertainty, which takes human knowledge into if-then rules and analyses the reasoning process. But, it is short of accurate quantitative analysis. In this research, we used the Adaptive Neural-Based Fuzzy Inference System (ANFIS) to handle the nitrate concentration data. ANFIS integrates the advantages of ANN and FIS models. Furthermore, the ANFIS model employs fuzzy if-then rules, can model the qualitative aspects

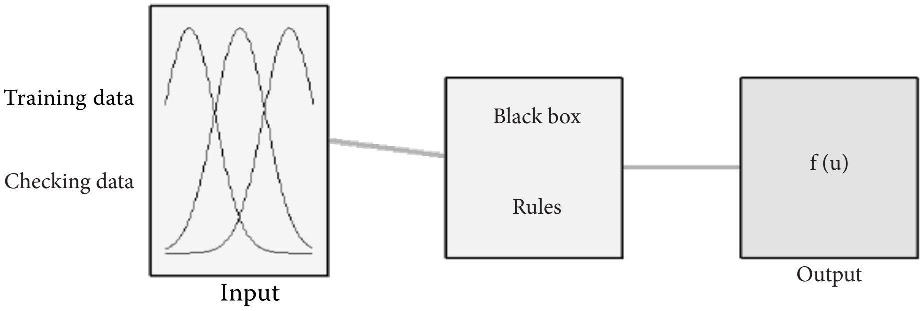


Figure 2. Fuzzy Inference System (FIS)

of human knowledge, and deals with uncertainty and nonlinear problems. In this way, one can bring the low-level learning and computational power of neural networks to fuzzy control systems and also provide the high-level reasoning of fuzzy control systems to neural networks (Cheng *et al.* 2009).

Various types of fuzzy inference system (FIS) are studied in the literature, and each one is characterized by consequent parameters (Figure 2). In this section, a brief description of the principles of ANFIS model is presented. The reader is referred to Chang and Chang (2001) for more details. In this research, the ANFIS model was adopted from MATLAB software Version 7.6.

Fundamentally, ANFIS is a graphical network representation of Sugeno-type fuzzy systems, endowed by neural learning capabilities. The network is comprised of nodes with specific functions, or duties, collected in layers with specific functions (Tsoukalas & Uhrig 1997).

In order to illustrate ANFIS's strength, the neural fuzzy control systems are considered based on the Tagak-Sugeno-Kang (TSK) fuzzy rules, whose consequent parts are linear combinations of their preconditions. The TSK fuzzy rules are in the following forms:

R^j: if x_1 is A_1^j and x_2 is A_2^j and .... and x_n is A_n^j,

then

$$y = f_j = a_0^j + a_1^j x_1 + a_2^j x_2 + \ldots + a_n^j x_n \tag{1}$$

where:

x_i (i = 1, 2, ..., n) – input variables

y – output variable

A_i^j – linguistic terms of the precondition part with membership functions $\mu A_i^j(x_i)$

$a_1^j \ldots a_1^j \in R$ (j = 1, 2, ..., n) – coefficients of linear equations f_j (x_1, x_2, ..., x_n)

To simplify the discussion, it is necessary to focus on a specific neurofuzzy controller (NFC) of the ANFIS type as an example.

Let us assume that the fuzzy control system under consideration consists of two inputs x_1 and x_2 and one output y and that the rule base contains only two TSK fuzzy rules as follows:

R^1: if x_1 is A_1^1 and x_2 is A_2^1, then

$$y = f_1 = a_0^1 + a_1^1 x_1 + a_2^1 x_2 \tag{2}$$

R^2: if x_1 is A_1^2 and x_2 is A_2^2, then

$$y = f_2 = a_0^2 + a_1^2 x_1 + a_2^2 x_2 \tag{3}$$

In the TSK fuzzy system, for given input values x_1 and x_2, the inferred output y^* is calculated by the following formula:

$$y^* = \frac{(\mu_1 f_1 + \mu_2 f_2)}{\mu_1 + \mu_2} \tag{4}$$

where:

μ_j – firing strengths of R^j (j = 1, 2)

given by the equation:

$$\mu_j = \mu_{A_1^j}(x_1) + \mu_{A_2^j}(x_2),\ j = 1, 2 \tag{5}$$

If the product inference is used, the corresponding ANFIS architecture is shown in Figure 3.

Node functions in the same layer are of the type described below. This is an ANFIS architecture where the following meanings can be attached to each layer:

Layer 1: Every node in this layer implies an input and it just passes external signals to the next layer.

Layer 2: Every node in this layer acts as a membership function $\mu_{A_1^j}(x_i)$ and its output specifies the degree to which the given x_i satisfies the quantifier A_i^j Generally, $\mu_{A_1^j}(x_i)$ is selected as

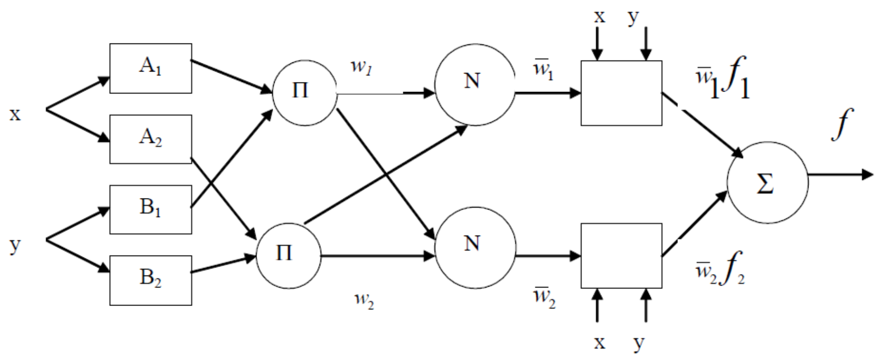


Figure 3. Structure of ANFIS

bell-shaped with a maximum equal to 1 and minimum equal to 0, so that:

$$\mu_{A_i^j}(x_i) = \frac{1}{\left(1 + \left\{\left[\frac{(x_i - m_i^j)}{\sigma_i^j}\right]^2\right\}^{b_i^j}\right)} \tag{6}$$

or:

$$\mu_{A_i^j}(x_i) = \exp\left\{-\left[\left(\frac{(x_i - m_i^j)}{\sigma_i^j}\right)^2\right]^{b_i^j}\right\} \tag{7}$$

where:

$\{m_i^j, \sigma_i^j, b_i^j\}$ – parameter set to be tuned

In fact, continuous and piecewise differentiable functions, such as commonly used trapezoidal or triangular membership functions, are also qualified candidates for node functions in this layer. Parameters in this layer are referred to as precondition parameters.

Layer 3: Every node in this layer is labelled Π and multiplies the incoming signals $\mu_j = \mu_{A_1^j}(x_1) + \mu_{A_2^j}(x_2)$ and sends the product out. Each node output represents the firing strength of a rule.

Layer 4: Every node in this layer is labelled N and calculates the normalized firing strength of a rule. That is the j^{th} node calculates the ratio of the firing strength of the jth rule to that of all the rules as follows:

$$\bar{\mu}_j = \mu_j/[\mu_{A_1^j}(x_1) + \mu_{A_2^j}(x_2)] \tag{8}$$

Layer 5: Every node j in this layer calculates the weighted consequent value as follows:

$$\bar{\mu}_j\,(a_0^j + a_1^j x_1 + a_2^j x_2) \tag{9}$$

where:

$\bar{\mu}_j$ – output of Layer 4

(a_0^j, a_1^j, a_2^j) – set to be tuned

Parameters in this layer are referred to as consequent parameters.

Layer 6: The only node in this layer is labelled Σ, and it sums all incoming signals to obtain the final inferred result for the whole system (Lin & Lee 1996).

A flow diagram for running ANFIS is shown in Figure 4.

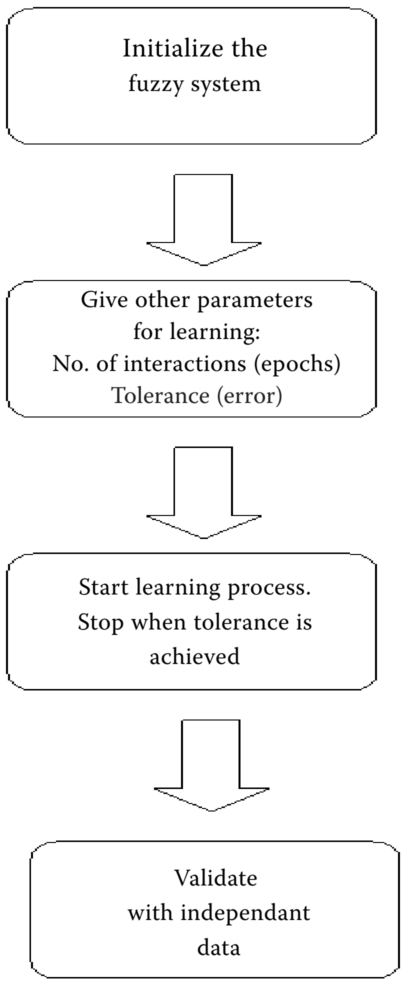


Figure 4. Flowchart of computations in ANFIS

Application

To evaluate the performance of ANFIS model in nitrate estimation, two performance criteria were used, namely the root mean square error (RMSE) and the determination coefficient (R^2). These criteria were defined by WILLMOTT *et al.* (1985) and ZACHARIAS *et al.* (1996):

$$\mathrm{RMSE} = \sqrt{\frac{1}{N}\sum_{i=1}^{n}\left(X_k - Y_k\right)^2} \tag{10}$$

$$R^2 = \left[\frac{\sum_{K=1}^{n}\left(X_k - \bar{X}\right)\left(Y_k - \bar{Y}\right)}{\sum_{K=1}^{n}\left(X_k - \bar{X}\right)^2 \sum_{Y=1}^{N}\left(Y - \bar{Y}\right)^2}\right]^2 \tag{11}$$

where:
X_k – measured value
Y_k – predicted value
$\bar{X}$ – mean of observed values
$\bar{Y}$ – mean of predicted values

The linear regression was applied between the measured (Y) and predicted (X) values of nitrate as follows:

$$Y = pX + q \tag{12}$$

where:
p – slope of line
q – intercept

If the value of q is not significant at a 5% level, it is considered zero.

RESULTS AND DISCUSSION

The applicability of ANFIS method was investigated to predict the average nitrate concentrations in 175 observation wells in Isfahan province. The groundwater quality in the observation wells was previously described in detail by JAFARI MALEKABADI (2002). In the present study, easily measurable water quality parameters (Table 1) were used in the prediction of nitrate concentrations. The data set (5-month averages) was divided into two groups: 122 observations (70% of the data set) for building the model (training data set) and 53 observations (30% of the data set) for validating the model (validation data set). This selection was done randomly. The number of membership functions for each input of ANFIS was set to 3 (each variable may have several values /in terms of rules/ and each rule includes several parameters of membership functions). For instance, for 4 variables, if each variable has 3 rules and each rule includes 3 parameters, then there are 36 (4 variables × 3 rules × 3 parameters) parameters to be determined in Layer 2. The only reason for having 3 memberships for each variable is the reduction of the number of rule-based alternatives.

In order to suit the consistency of the model, all source data were first normalized in the range from 0.0 to 1.0 and then returned to original values after the simulation, using:

$$x_{\mathrm{norm}} = \frac{\left(x_i - x_{\min}\right)}{\left(x_{\max} - x_{\min}\right)} \tag{13}$$

Table 1. Significance of regression between nitrate concentration and input variables

Variable	Regression coefficient	Min	Max	Mean
pH	0.06 ns	7.45	8.73	8.05
SAR	0.009 ns	0.53	148.96	7.029
SO_4	0.047 ns	1.12	64.12	11.89
Na	0.036 ns	19.4	7042.5	445.58
K	0.081*	0.78	54.88	5.91
Cl	0.1**	1.4	287.75	18.64
Hardness	0.35***	180	3091	680.24
EC	0.278***	0.33	25.92	3.14
Mg	0.29***	0.84	30.66	5.902
Ca	0.319***	0.73	31.16	4.166
HCO_3	0.266***	2.3	7.9	3.87

*significant at 5% level, ** significant at 1% level, *** significant at 0.1% level; ns – not significant

Table 2. Number of structures for determination of the best input data

No. of input data	No. of structures	The best input data (based on the highest R^2 and smallest RMSE)
5	21	HCO_3, Ca, Mg, hardness and EC
4	35	HCO_3, Ca, hardness and EC
3	35	hardness, EC and HCO_3
2	21	hardness and EC

where:

x_{norm} – standardized data

x_{max}, x_{min} – maximum and minimum measurement values

Such standardization procedure renders the data into a dimensionless form.

We had 11 data sets of information. Among the 11 water quality variables considered (K, Mg, Na, Cl, HCO_3, SO_4, Ca, hardness, pH, EC and SAR), it was clear that some would play a more important role than others and it was important that only the significant ones would be used as inputs for the final model. Therefore, different combinations of the data were created to build the ANFIS model. First, we used the relationships between input variables and nitrate concentration. Then, the non-significant parameters (pH, SAR, SO_4, and Na) were removed from the list.

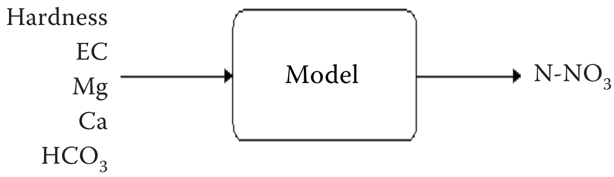
Different methods exist for selecting the best input data set. For example, Keskin *et al.* (2004) suggested that using the statistical analysis according to correlation coefficients selects the best input data set. Kisi (2005) reported that a non-linear method instead of correlation analysis should be for determination of the degree of effectiveness between the output and each input parameter. In this method, different input combinations could be tried using fuzzy models in a non-linear manner, in order to choose the best one.

In this paper, Keskin *et al.* (2004) method was used. Table 1 shows the performance and the statistical coefficients of input variables vs. nitrate concentration. Based on these results, the four parameters of pH, SAR, SO_4 and Na were removed from a further comparison analysis.

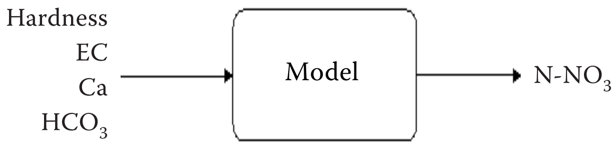
To determine the most suitable network, different numbers of parameters (out of seven parameters of K, Cl, hardness, EC, Mg, Ca and HCO_3) were considered. In this respect, for 5 input data, 21 structures were constructed; for 4 input data, 35 structures; for 3 input data, 35 structures; for 2 input data, 21 structures. For each of the input sets, the structure with the smallest RMSE and the highest R^2 was selected as the best. The final results of different structures are shown in Table 2. It should be mentioned that the K and Cl parameters were not in the best network with different inputs. Therefore, five parameters of hardness, EC, Mg, Ca and HCO_3 are the parameters which are used in the main body of the research.

Based on the input data, 4 ANFIS models were obtained as follows:

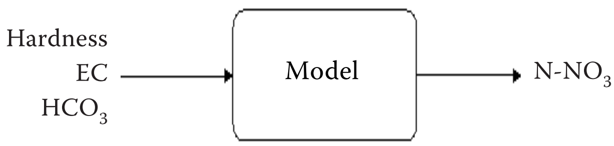
ANFIS1



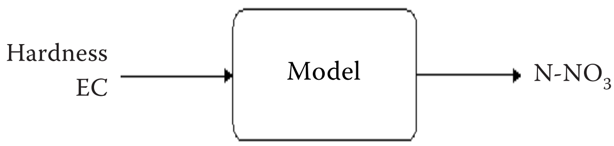
ANFIS2



ANFIS3



ANFIS4



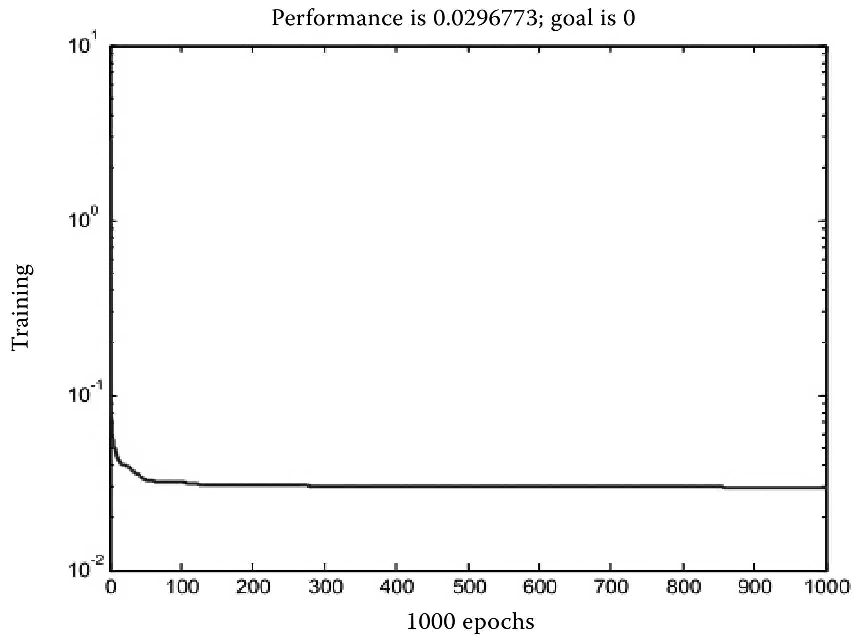


Figure 5. Plot of MSE vs. number of epochs for training data in ANFIS1

Table 3. Regression equation of different models compared to measured nitrate

Model	Input	Equation	R^2	RMSE	Performance
ANFIS1	EC, HCO_3, Ca, Mg, hardness	$N_{measured} = 1.07\ N_{ANFIS1}$	0.93	1.17	very good
ANFIS2	EC, HCO_3, Ca, hardness	$N_{measured} = 1.17\ N_{ANFIS2}$	0.91	1.9	good
ANFIS3	EC, HCO_3, hardness	$N_{measured} = 1.2\ N_{ANFIS3}$	0.88	2.3	reasonable
ANFIS4	EC, hardness	$N_{measured} = 1.25\ N_{ANFIS4}$	0.68	2.94	not good

For all the above four ANFIS models, the number of epochs were altered and the corresponding MSE was obtained. For example, Figure 5 demonstrates the training progress of ANFIS1 model with time. This figure shows the downward trend of MSE with increasing number of iterations. It can be seen that approximately after 1000 epochs, the MSE for the training data set is less than 0.03.

Using the input data in the above models, the nitrate concentration was obtained. The performance and

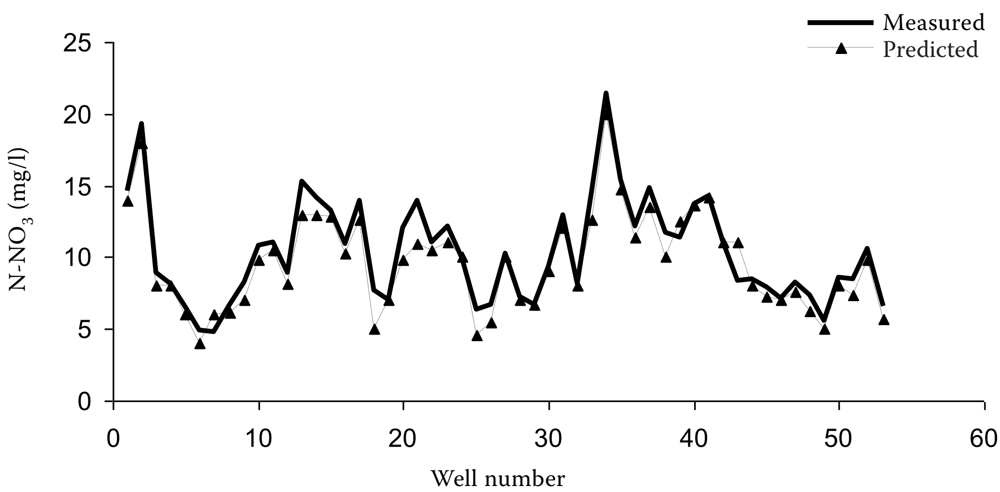


Figure 6. Predicted average nitrate concentration using ANFIS1

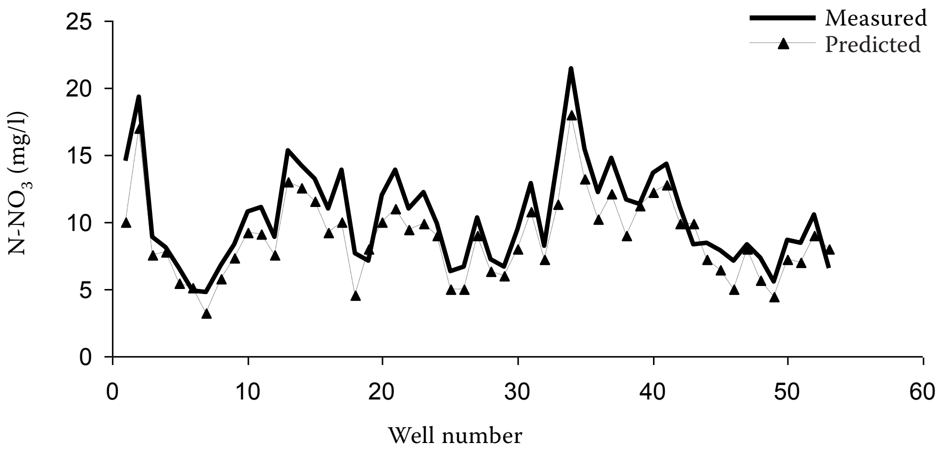


Figure 7. Predicted average nitrate concentration using ANFIS2

the statistical coefficients obtained for each model are shown in Table 3. According to this table, ANFIS1 provided the best estimates of NO_3 and resulted in the lower value of RMSE (1.17), followed by ANFIS2 (1.9), ANFIS3 (2.3) and ANFIS4 (2.94). The NO_3 values of ANFIS1 were lower than the measured NO_3 (Figure 6), where the ratio $N_{measured}/N_{ANFIS1}$ is 1.07. ANFIS1 underestimated NO_3 by 7%. A possible reason for the better performance by ANFIS1 model may be that it has a higher number of input parameters with respect to other models (Table 3).

The performance of ANFIS2 was also good (Figure 7), with RMSE = 1.9 and R^2 = 0.91. The performance of ANFIS3 showed that this model can provide a reasonable estimation of NO_3 (Figure 8). Results of ANFIS4 (Figure 9) showed that this model cannot provide a good estimation of NO_3. ANFIS2 and ANFIS3 models underestimated the measured NO_3 by 17% and 20%, respectively.

The results are in a good agreement with Terzi *et al.* (2006) and Keskin *et al.* (2009), who showed that ANFIS model can compete with direct methods for the estimation of parameters. These results are consistent with the performance of ANFIS reported by Jang (1992) and Nayak *et al.* (2004), who showed that this model was used successfully in many applications. Results in the literature (Ramasamy *et al.* 2003; Sacco *et al.* 2007) documented that regression and neural networks were used to predict the nitrate concentration in groundwater. But such approaches are rather scarce in the literature.

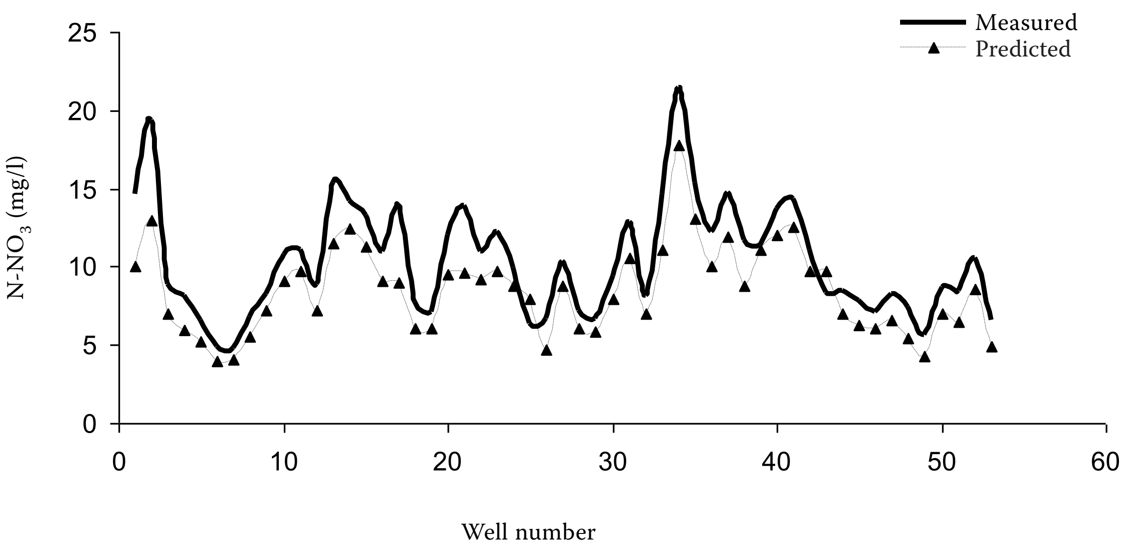


Figure 8. Predicted average nitrate concentration using ANFIS3

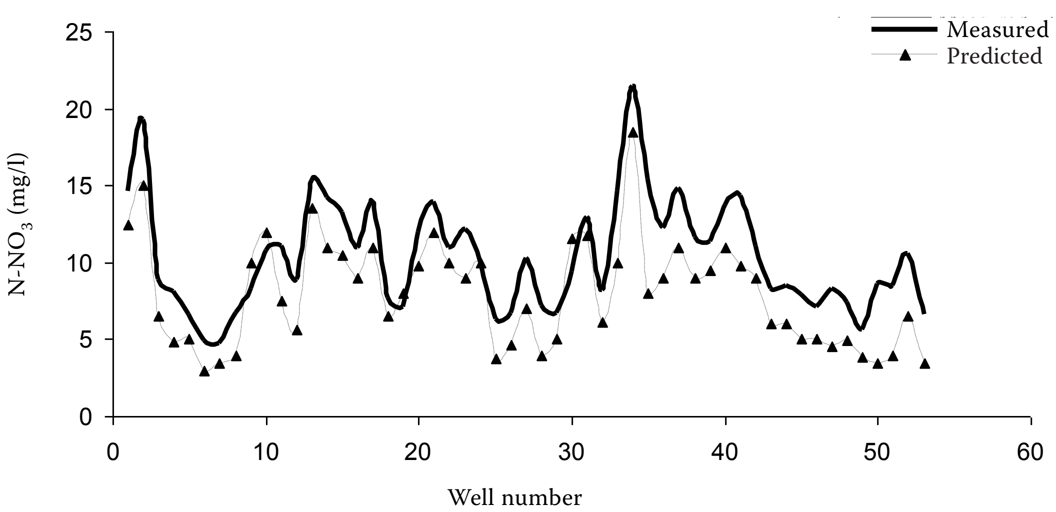


Figure 9. Predicted average nitrate concentration using ANFIS4

CONCLUSION

Public concern over the deterioration of groundwater quality from nitrate contamination has grown significantly in recent years. This concern has focused increasingly on anthropogenic sources. In this study, the suitability of ANFIS model was examined for estimating the average NO_3^- concentration by means of some measured groundwater data from 175 wells in Isfahan province, Iran. Eleven water quality variables including the average K, Mg, Ca, Na, Cl, HCO_3, SO_4, hardness, pH, EC and SAR were considered, out of which 5 variables, EC, HCO_3, Ca, Mg and hardness, were selected as the most effective. The results showed that an increase in the number of input variables in ANFIS models improves the accuracy of nitrate estimates. The ANFIS1 model provided the best estimates of NO_3^- with the lowest RMSE and the highest R^2 values, followed by ANFIS2, ANFIS3 and ANFIS4 models. To find the best model for nitrate estimation, it is recommended that those parameters which have the highest correlation be chosen and those which have the least correlation be discarded. In general, the results of the current research could be useful for groundwater management purposes. The findings of this research could be applied in practice for the indirect monitoring of nitrate concentration where some other chemical data are available from the wells. This saves time and expenses.

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