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Application of Geostatistical Methods for determining nitrate concentrations in Groundwater (case study of Mashhad plain, Iran)

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ABSTRACT: Excess nitrate in the soil can be easily moved with Irrigation water, rainfall or melting snow to pass through different layers of soil and reach groundwater. Water pollution occurs when the elements in the water exceeds the limit. If nitrate in drinking water is greater than the limit it will cause disease in humans. The purpose of this study was the evaluation of geostatistical methods for estimation of the spatial variables nitrate, sodium and electrical conductivity in groundwater of Mashhad (Iran). Samples were taken from 276 wells and the parameters nitrate, sodium and electrical conductivity were measured. Data were analyzed with software GS+. To investigate the spatial correlation of data, experimental Variogram each variable were calculated and plotted. Amounts of nitrates, sodium and electrical conductivity were estimated with using geostatistical methods. Cross validation method was used to determine the accuracy of the estimated data. After drawing the variogram, the best model was fitted based on minimum RSS. For determining the most appropriate method estimation of the root mean square error (RMSE) and scatter plot method was used. Results showed that kriging methods are more accurate. Sodium and nitrate maps were drawn based on kriging method and areas where nitrate concentrations greater than 50 mg per liter, were identified. Comparing the plans of sodium and nitrate showed that sodium and nitrate concentrations in a certain part of the scope of this study are higher than the limit.

***Keywords**:* Geostatistical, Interpolation, Kriging, nitrate, water quality, sodium

INTRODUCTION

Water is essential for sustenance of life. Groundwater is used for domestic and industrial water supply and irrigation all over the world. In the last few decades, there has been a tremendous increase in the demand for fresh water due to rapid growth of population and the accelerated pace of industrialization (Mevlut at el., 2010). Human health is threatened by most of the agricultural development activities particularly in relation to excessive application of fertilizer and unsanitary conditions. (Amadi., 2010) . Groundwater is the most important resources used for drinking and utility and irrigation purposes in this region. Most of drinking water sources has been contaminated by microbial or chemical contaminants potentially hazardous to human health (Sylvestre and Rodriguez., 2008) on the part of intensive agriculture, answered sanitation in densely populated areas, or from point sources such as irrigation of land by sewage effluents (Sutharet al., 2009). Nitrate contamination in groundwater is a common problem in many part of the world (Suthar et al., 2009; Almasri., 2007). Nitrate is naturally occurring ions that are part of the nitrogen cycle. The nitrate ion is the stable form of combined nitrogen for oxygenated systems. Although chemically un reactive, it can be reduced by microbial action (Nas and Berktay., 2006). The World Health Organization (WHO) and the European Community (EC), maximum contaminant level (MCL) of nitrate is given to be 50 mg/L. High levels of nitrate in drinking water can cause illness in humans. Nitrate increases the risk of cancer in humans.

Today, the nitrate contamination of groundwater in some parts of Iran is serious. Nitrate contamination of groundwater in agricultural areas where excessive use of nitrogen fertilizer.

Water resources planning and management provide decision-tools for :
1. allocation of adequate water to the consumers at appropriate time and place
2. protection from excessive water (floodwater)
3. maintenance of acceptable water quality (Loucks., 1981).

Groundwater quality mapping over extensive areas is the first step in water resources planning (Todd., 1980). One of tools for mapping of groundwater quality is geostatistical methods. Geostatistical methods were developed to create mathematical models of spatial correlation structures with a variogram as the quantitative measure of spatial correlation. The variogram is commonly used in geostatistics and the interpolation technique, known as kriging, provides the ‘‘best’’, unbiased, linear estimate of a regionalized variable in an unsampled location, where ‘‘best’’ is defined in a least-squares sense. The emphasis is set on local accuracy, closeness of the estimate to the actual, but unknown, value without any regard for the global statistical properties of the estimates. The kriging estimation variances are independent of the value being estimated and are related only to the spatial arrangement of the sample data and to the model variogram (Webster and Oliver., 2001). Geostatistics provides a set of statistical tools for analyzing spatial variability and spatial interpolation. These techniques produce not only prediction surfaces but also error or uncertainty surfaces. A semivariogram is used to describe the structure of spatial variability. The semivariogram plays a central role in the analysis of geostatistical data using the kriging method. They take into account the spatial autocorrelation in data to create mathematical models of spatial correlation structures commonly expressed by variograms (Gundogdu and Guney., 2007). A major advantage of geostatistic (kriging) is that it is more flexible than other methods used interpolation and spatial averaging such as inverse distance weighing and deterministic splines, which are used in interpolation and contouring, or thissen polygons. Another advantage of geostatistic is that it provides the means to evaluate the magnitude of the estimation error. The mean square error is a useful rational measure of the reliability of the estimate. It depends only on the variogram and the location of the measurements (Kitanidis., 1996). Marinoni (2003) applies a combination of ordinary and indicator Kriging, in order to reduce the smoothing error of the estimated regionalised function. A number of very interesting applications concern the use of Kriging with external drift. In their work Merz and Bloschl (2004) apply Kriging with external drift (using elevation as additional information) for the estimation of the distribution of precipitation, air temperature and soil evapo-transpiration. Desbarats et al. (2002) extend this application to aquifer systems using the assumption that ‘‘water table in phreatic aquifers is a subdued replica of the ground surface above’’, in order to increase accuracy in the estimation of groundwater level distribution. In order to achieve that, they express water table in terms of a deterministic trend given by topographic elevation and a component representing depth to water. Amini et al. (2002) applied kriging and co-kriging techniques for predicting Cl^- concentration of soil in Isfahan which showed that kinging method provides more accurate and low cost results. Jager et al. (1990) also used geostatistical tools like kriging to simulate groundwater quality variables and who added result kriging is better than other geostatistical tools for simulating groundwater quality variables. Misaghi and Mohammadi (2002) estimated groundwater table by using geostatistics methods. Nazari et al. (2006), used geostatistics method to study spatial variability of groundwater quality in Balarood plain. Their results showed that spherical model is the best model for fitting an experimental variograme of EC, Cl and SO4 variables. Istock and Cooper (1998) used kriging to estimate heavy metals concentration in groudwater and concluded that it the mentioned method is the best estimator for spatial prediction of lead. Dagostino et al. (1998) studied spatial and temporal variability of groundwater nitrate, using kriging and cokriging methods. Their results show that cokriging method has higher accuracy than kriging in estimating of nitrate concentration. Rizzo and Mouser (2000) used geostatistics for analyzing groundwater quality. They used microbial data as an auxiliary variable in cokriging method. Their results show that cokriging method has suitable accuracy to estimate groundwater quality. Ahmad (2002) found that kriging method has a high accuracy in estimating of total dissolved salts (TDS) in groundwater. Gaus et al. (2003) studied groundwater pollution in Bangladesh. They used disjunctive kriging method to estimate arsenic concentration and to prepare risk map. Their results show that 35 milion people are exposed to high concentration of arsenic (50ppm).
Barca and Passarella (2007) used Disjunctive kriging and simulation methods to make nitrate risk map in 10, 50(mg/l) thresholds, in Modena plain of Italy. Their results showed that Disjunctive kriging method is the suitable method to study deterioration level of Groundwater. Because of various results reported by above mentioned researchers, it is obvious that suitable method of interpolation to estimate one variable depends on variable type and regional factors, thus any selected method for specific region cannot be generalized to others.

This research has been carried out with the aim of testing the performance of spatial interpolation techniques for mapping Groundwater quality and also evaluation geostatistical methods for estimation of the spatial variables nitrate, sodium and electrical conductivity in groundwater of Mashhad (Iran).

MATERIAL AND METHODS

geostatistics

Geoscientists often face interpolation and estimation problems when analyzing sparse data from field observations. Geostatistics is an invaluable tool that can be used to characterize spatial or temporal henomena.

Geostatistics originated from the mining and petroleum industries, starting with the work by Danie Krige in the 1950's and was further developed by Georges Matheron in the 1960's. Geostatistics has since been extended to many other fields in or related to the earth sciences, e.g., hydrogeology, hydrology, meteorology, oceanography, geochemistry, geography, soil sciences, forestry, landscape ecology. In this class, both fundamental development of geostatistics and simple, practical applications in the earth sciences will be presented. Advantage of geostatistics is the use of quantitative measures of spatial correlation, commonly expressed by variograms (Diodato and Ceccarelli., 2005). The semivariogram is a fundamental tool in geostatistics.

The basic principles of Kriging

Kriging is a method for linear optimum unbiased interpolation with a minimum mean interpolation error. Kriging is known to be an exact estimator in the sense that observation points are correctly re-estimated. The method does not necessarily require observation networks where data are normally distributed and for the estimation of the structure of the regionalised variables it takes into consideration only the neighbouring points of estimation data. The term ‘‘structure’’ refers to the spatial correlation of the variable in different points of the area under study (de Marsily., 1986). One of the main advantages of Kriging, although there is no general consensus on its usefulness, is that it presents the possibility of estimation of the interpolation error of the values of the regionalised variable where there are no initial measurements. This feature offers a measure of the estimation accuracy and reliability of the spatial distribution of the variable. The spatial variability of a regionalised variable is described by a semi-variogram. The empirical semivariogram is a graphical representation of the mean square variability between two neighbouring points of distance h as shown in Eq. (1),

$$\gamma(h)=\frac{1}{2n(h)}\sum_{i=1}^{n(h)}\left[(z(x+h)-z(x))^2\right] \quad (1)$$

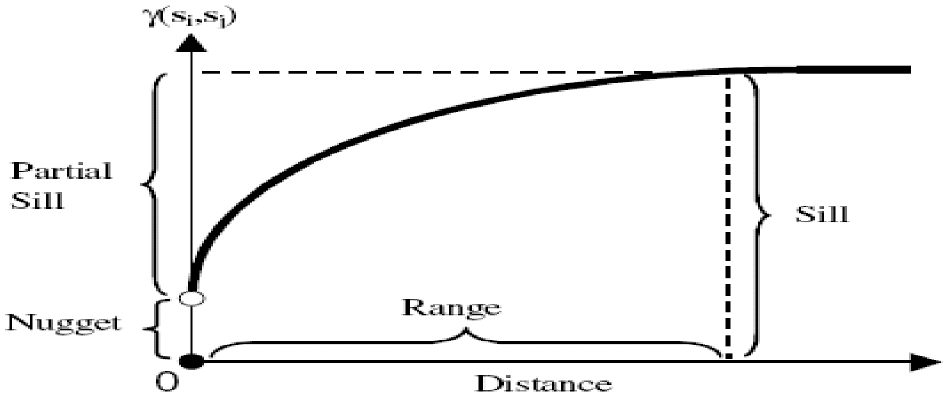

Figure 1. semivariogram

where z(x) and z(x+h) are the values of the variable at point x and at a point of distance h from point x. At the empirical semi-variogram, a theoretical one is adjusted whose equation is derived based on the principle that Kriging, as an exact estimator, re-evaluates correctly the measurements at the observation points (de Marsily., 1986; Journel., 1989). The empirical semivariogram (γ(h)) is defined as half the average quadratic difference between two observations of a variable separated by a distance vector h (Journel and Huijbregts., 1978).

Kriging is an interpolation method used in a large number of applications concerning a wide range of different fields. One of the main problems regarding the application of any interpolation technique concerns the imprecision of the original data. Before the geostatistical estimation, a variogram is calculated for classes of distance between sample pairs. The most widely used models are spherical, exponential, Gaussian and pure nugget effect (Isaaks and Srivastava., 1989). The validation and the sufficiency of the developed model variogram can be tested via a technique called cross validation. Cross validation estimation is obtained by leaving one sample out and using the remaining data. This test allows assessing the goodness of fitting of the variogram model, the appropriateness of neighbourhood and type of kriging used. The interpolation values are compared to the real values and then the least square error models are selected for regional estimation (Leuangthong et al., 2004). Kriging is a method for linear optimum appropriate interpolation with a minimum mean square error. One of the main advantages of Kriging, although there is no general consensus on its usefulness, is that it presents the possibility of interpolation estimation error of the regionalised variable (ReV) where there are no initial measurements. This feature offers a measure of the estimation precision and reliability of the spatial variable distribution (Theodossiou and Latinopoulos, 2006). The kriging estimator is given by a linear combination. The general equation of the kriging method is as follows:

$$\hat{Z}(x_0) = \sum_{i=1}^{n} w_i(x_0) Z(x_i) \qquad (2)$$

of the observed values $z_i = Z(x_i)$ with weights $w_i(x_0),\ i = 1, \ldots, n$

The model parameters (range and sill variance) describe the structure of spatial variation and are used for estimation at unsampled locations using kriging. A nugget variance parameter is common for sample data of a continuous variable. Measurement error and stochastic variation in data contribute to the nugget, the largest source of variation is commonly due to spatially dependent variation that occurs over distances much smaller than the shortest sampling interval. When the difference between samples in space is at a maximum for the average separation distances, the sill variance is reached and the model is bounded. The lag distance at which the variogram reaches its sill is the range, which indicates the limit of spatial dependence (Chappell et al., 2003).

Validation estimates

The accuracy and validity of the method was estimated using cross - validation. To compare observed and predicted values, there are a number of criteria. The most importants are:

Mean Absolute Error (MAE)

$$\mathrm{MAE} = \frac{1}{n}\sum_{i=1}^{n}\left|z^*(x_i) - z(x_i)\right|$$

Mean Bias Error (MBE)

$$\mathrm{MBE} = \frac{1}{n}\sum_{i=1}^{n}\left(z^*(x_i) - z(x_i)\right)$$

Root Mean Square Error(RMSE)

$$\mathrm{RMSE} = \frac{1}{n}\sqrt{\sum_{i=1}^{n}(z^*(x_i) - z(x_i))^2}$$

In these equations: Z* (xi) Estimated value and Z (xi) Measured value.

scatter plot of observed and predicted values

In this method, the estimated and observed values are drawn towards each other. The distribution amount is closer to the 45 degree line, indicating that the method has a more precise estimate. If the estimated values are equal to observational data, points fall exactly on the line of 45 degrees. More distribution points around the 45 degree line, indicating that the large difference between observed and predicted values. If the concentration is below the 45 degree line (X axis are the observed values) indicate that the estimate the amount of the estimation lower and inverse (Saghafiyan et al., 2011).

Geographical location of the study area

The study area is located in the province of Khorasan. This area has been extended between the longitudes of 59o 20' to 60o 08' and latitudes 35o 40' to 36o 03' of Iran central plateau (Fig. 2).

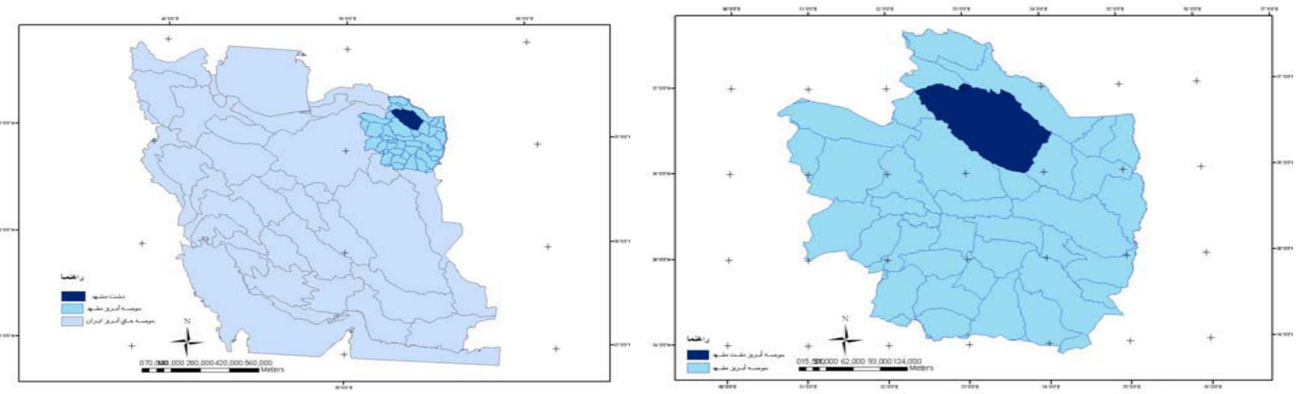

Figure 2. Location of study area

Sampling of water sources

For qualitative tests, the 276 drinking water wells were sampled. The wells were owned water and wastewater Mashhad. The samples were transported to the laboratory for testing. The electrical conductivity of the samples with EC meter and sodium by flame photometer were measured. Nitrate with Diazo method (De - Azo) and Cl with DPD method were determined . To determine the location of each well was read geographic coordinates with GPS devices.

RESULTS AND DISCUSSION

Normalization *of data*

One of the important things before choosing the appropriate method for the geostatistical is evaluation of data. Therefore, you should first check whether the data follow the normal procedure. If the data are not normal, it is necessary to use existing methods, the data are normalized. In this study, data Were examined with GS+ software, And then were normalized with logarithmic method. The following graphs show the trend of the normalized data.

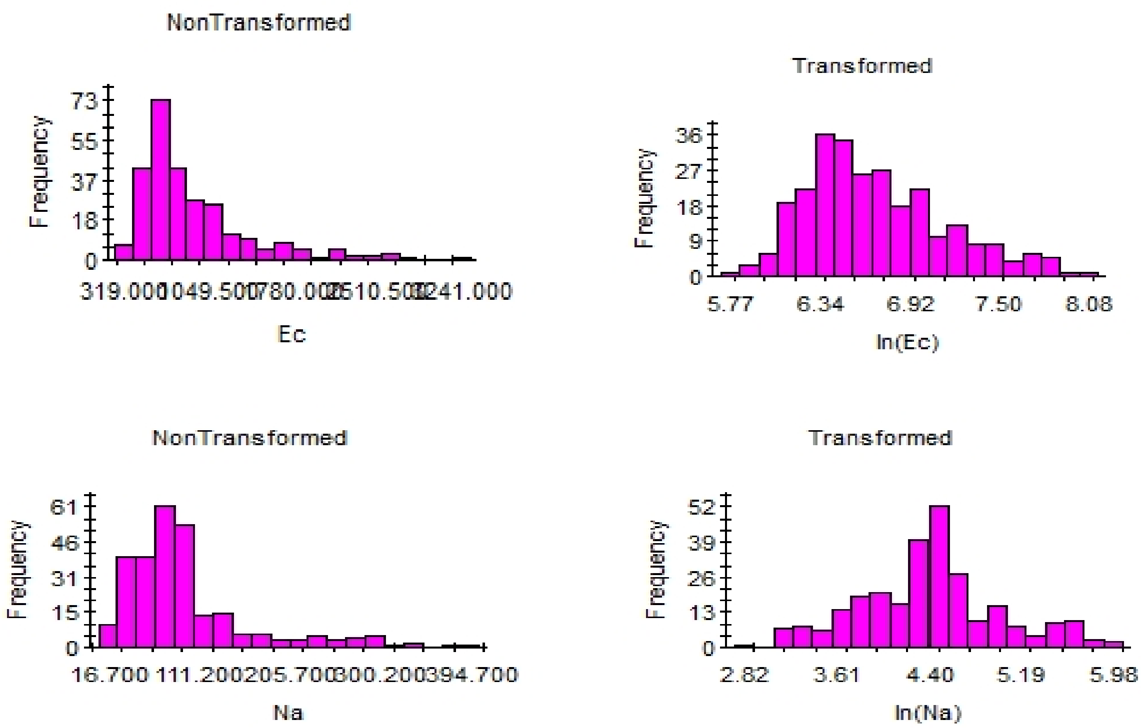

Figure 3 . Normalize the data

Variogram and spatial structure of data

To investigate the spatial correlation of data (presence or absence of spatial structure of the data) empirical variogram were calculated and plotted. The variograms of homogeneity of the environment, were drawn in different directions. The results showed that a strong association between parameters exist in all directions. After drawing variogram, models were fitted for them. The best fit of the model to the empirical variogram was chosen based on the minimum RSS. The results are given in Table 1.

Table 1 . Choose the model based on empirical variogram (RSS)

Gaussian	Exponential	Spherical	Quality parameters
0.715	** 0.685	0.693	Nitrate
0.242	** 0.233	0.235	Sodium (Na)
0.0695	** 0.0644	0.0720	Electrical conductivity (Ec)

** Selected models

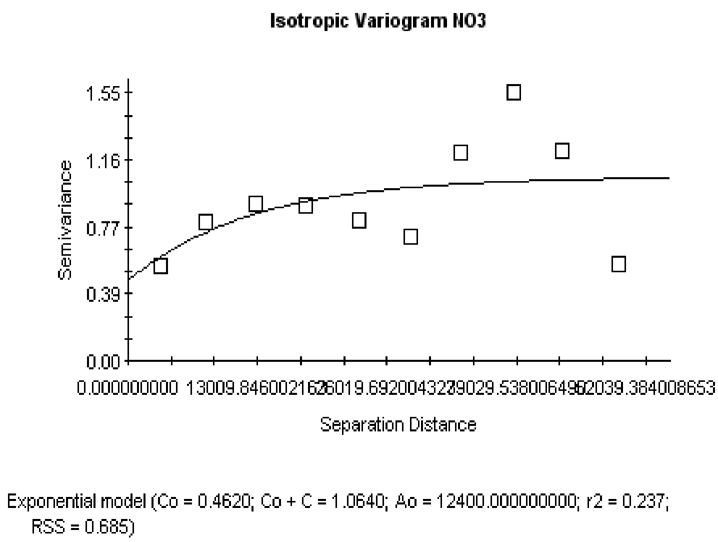

Figure 4. Nitrate Variogram

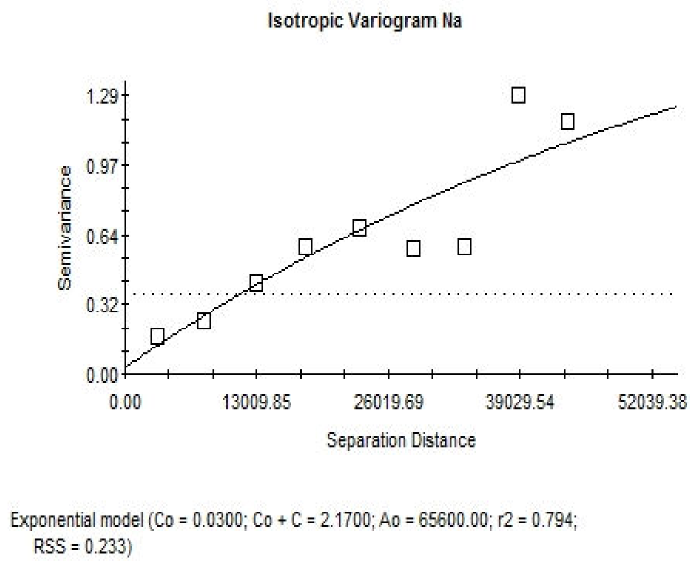

Figure5. Sodium Variogram

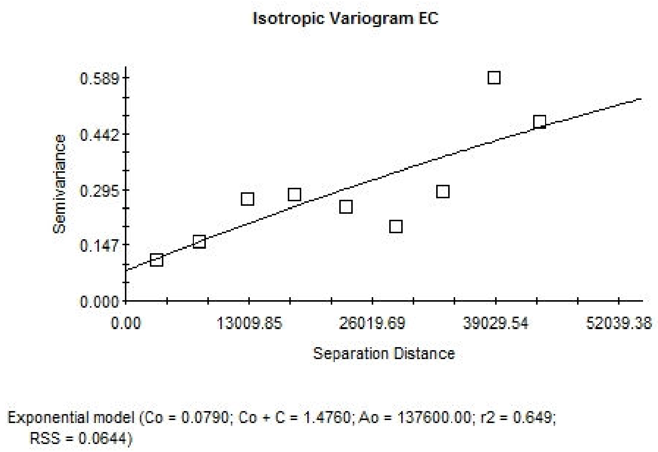

Figure 6. Ec Variogram

Variorums in different directions

Empirical variogram show spatial coherence of the data. It can also indicate the heterogeneity and anisotropy of the variable in the region. If the variogram sill and Radius of influence in different ways with different directions, the environment is anisotropy. After drawing the variogram in different directions, range effects and sill were relatively uniform. So anisotropy was not observed in the study area. Figure 7 shows the nitrate directional variogram. Nugget Effect to sill ratio is expressed as a percentage. This ratio is a measure of water quality parameters for classification of spatial dependence. If this value is less than 25%, the variable has strong spatial dependence. If this value is between 25 and 75%, the variable has moderate spatial dependence. If this value is greater than 75%, the variable has a weak spatial dependence. According to the results presented in Table 3, the parameters of sodium and electrical conductivity had the strong spatial structure. Also nitrate had medium spatial structure. Range of sodium, nitrate and electrical conductivity were 65, 12.4 and 137.6 Km, respectively.

Table 3 . Characteristics of experimental variogram was fitted

Parameter	Model	C_0 (Nugget)	(C_0+C)	A_0 (km) (Range)	$C_0/(C_0+C)$	R^2
Nitrate	Exponential	0.482	1.064	12400	0.45	0.227
Sodium (Na)	Exponential	0.03	2.17	65600	0.13	0.794
Electrical conductivity (Ec)	Exponential	0.079	1.476	137600	0.05	0.649

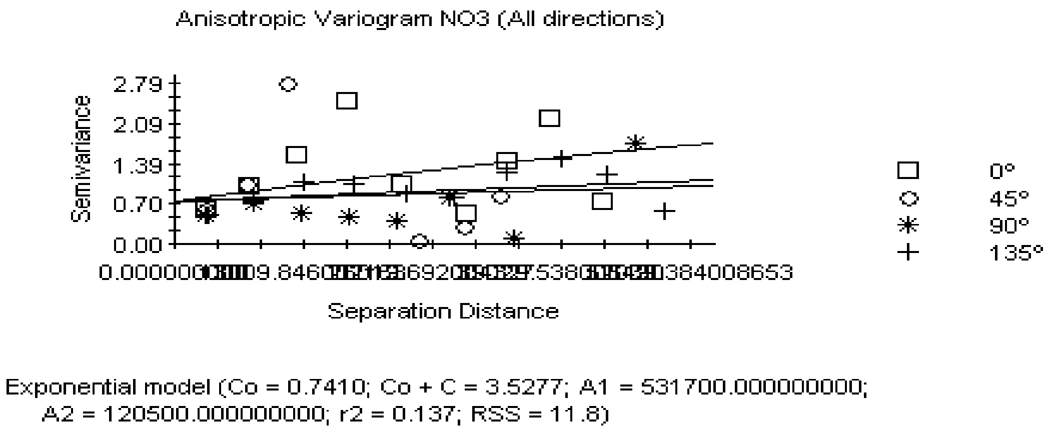

Figure 7. Directional variogram nitrate

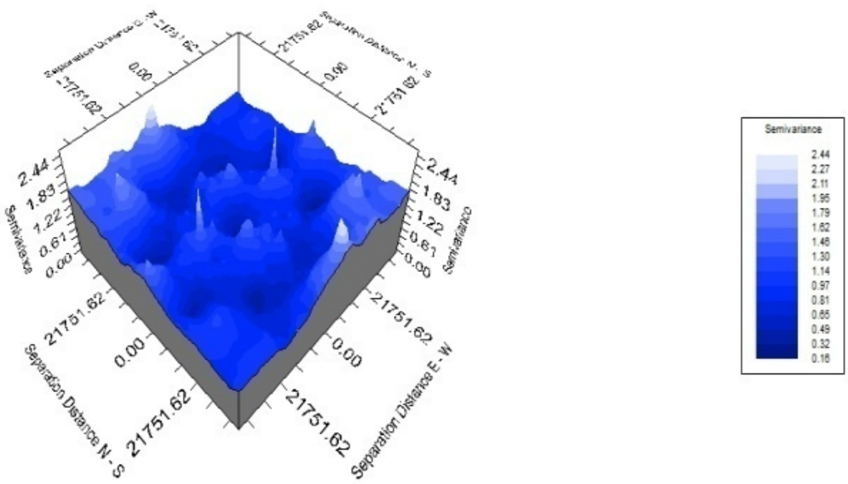

Figure 8 . Nitrate variogram in different directions

Validation of estimates

To determine the accuracy and validity of the data, the estimated nitrate, sodium and electrical conductivity of the cross-validation method was used. The results showed that kriging and inverse distance weight (IDW) methods estimated the amount of nitrates with good accuracy, although kriging method was more accurate (R2 = 0.544). kriging method estimated Sodium (R2 = 0.774) and electrical conductivity (R2 = 0.678) with greater precision. In general, kriging method had less estimation error and more precise estimation.

select the best method for estimating

For comparing Interpolation methods and select the best method from two methods, root mean square error (RMSE) and scatter plot were used. The results showed that the methods of geostatistical was more accurate than non geostatistical methods (inverse distance weight and normal distance weight). Table 4 shows values of RMSE and R2 (R2 is related to scatter plots).

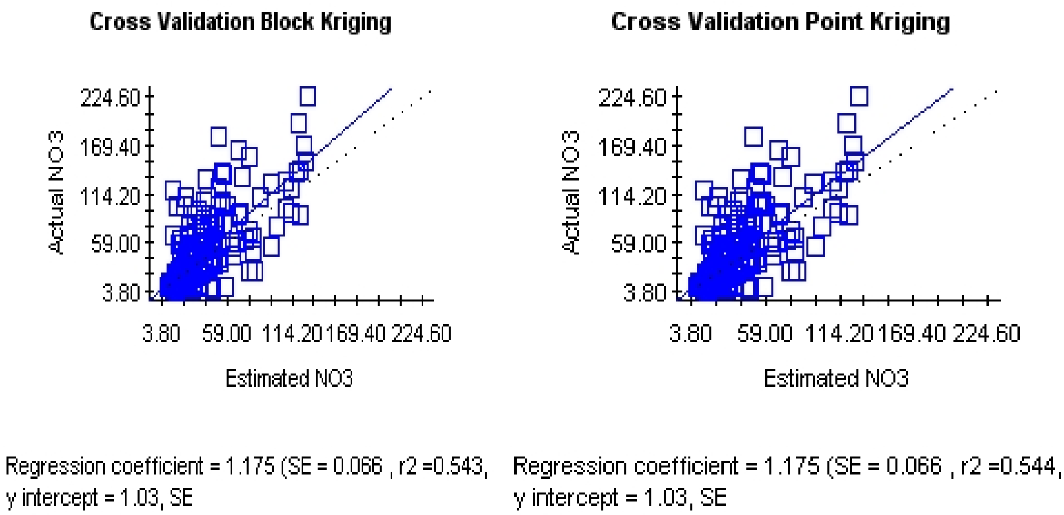

Figure 9 . Nitrate of Validation (block and point kriging)

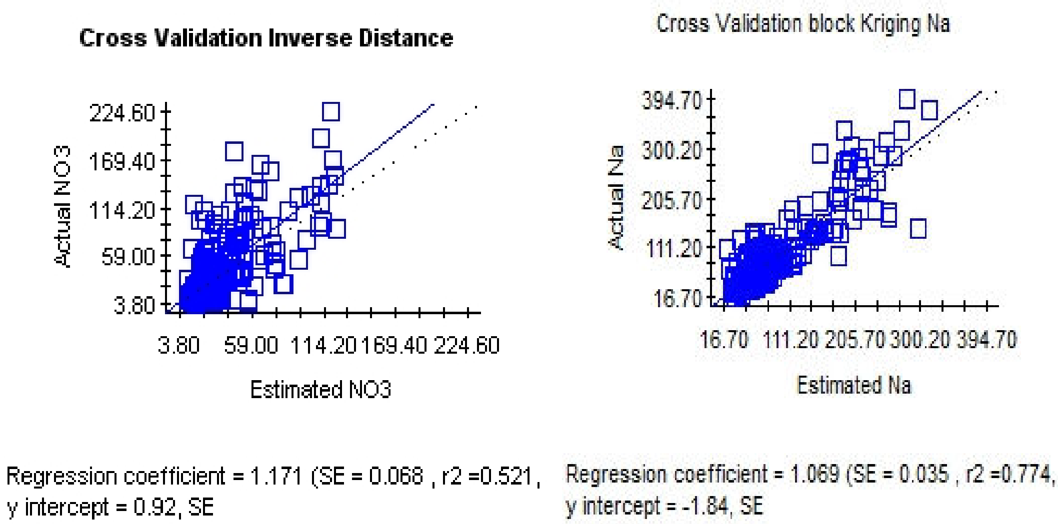

Figure 10. parametersof Validation

Table4. Select the appropriate interpolation method based on $R MSE$ and R^2

Parameter	NDW		IDW		Point Kriging		Block Kriging	
	R^2	RMSE	R^2	RMSE	R^2	RMSE	R^2	RMSE
Nitrate	0.398	0.080	0.5211	0.068	0.544	0.066	0.543	0.066
Sodium (Na)	0.524	0.068	0.669	0.050	0.774	0.035	0.774	0.035
Electrical conductivity (Ec	0.455	0.078	0.643	0.057	0.678	0.51	0.676	0.051

Draw maps

According to the results, kriging methods were more accurate. Sodium and nitrate zoning maps for the study area were drawn based on block kriging. (Fig. 11).

CONCLUSION

The main objective of this study was to determine the best estimators for providing nitrate, sodium and electrical conductivity maps in drinking water wells of Mashhad city. After selecting the appropriate method to estimate, the water quality maps of the area were drawn. Results of this study showed that, there is a strong spatial correlation between the studied parameters. When there is a strong spatial structure of the data, geostatistical methods are more accurate than the classical statistic methods. The results of this study are consistent with the results of Dagostino (1998), Jager(1990), Istock(1998), Ahmad(2002) and Barca(2007). After drawing the map of nitrates in the water, the areas where nitrate concentrations were higher than 50 mg per liter, were marked on the map. Removal of existing wells in this area were proposed. Also, the source of nitrate contamination in the study area should be investigated.

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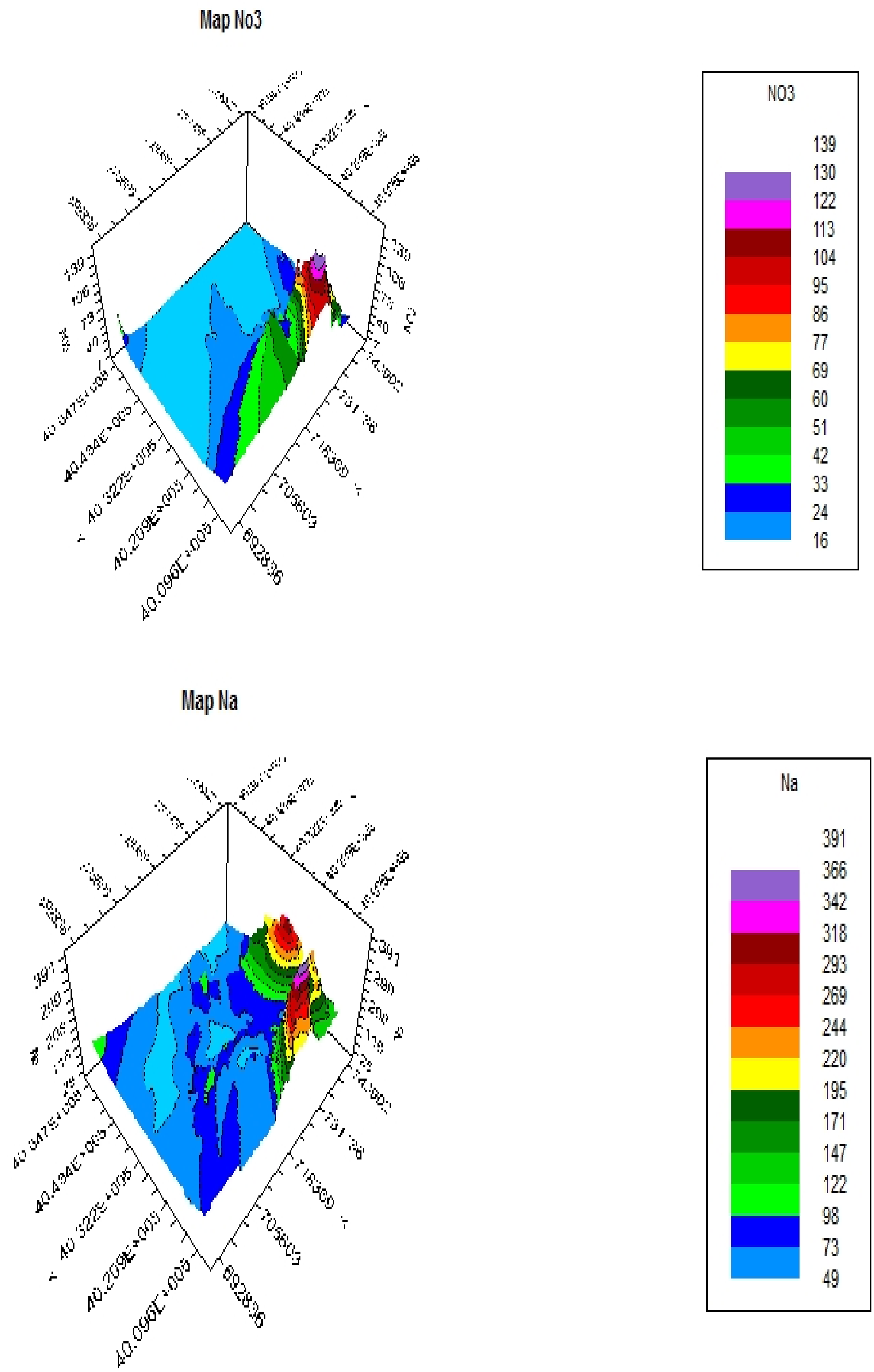

Figure 11. Nitrate and sodium of Maps (based on kriging)

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