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SPATIAL ANALYSIS OF THREE AGRICHEMICALS IN GROUNDWATER OF ISFAHAN USING GS+

***[1]M. M. Amin, [1]A. Ebrahimi, [2]M. Hajian, [3]N. Iranpanah, [1]B. Bina**

[1]Department of Environmental Health Engineering, School of Public Health, Isfahan University of Medical Sciences and Isfahan Research Center of Health of Environment, Isfahan, Iran
[2]Department of Civil Engineering, School of Engineering, Isfahan University, Isfahan, Iran
[3]Department of Statistics, School of Mathematics and Statistics, Isfahan University, Isfahan, Iran

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ABSTRACT
The purpose of this study was to undertake a spatial analysis of total organic carbon, electrical conductivity and nitrate, in order to produce a pollution dispersion and prediction map for the investigated area in the province of Isfahan in Iran. The groundwater samples were collected from a zone as a pilot study area of 80 km^2, including 25 water wells, based on the criteria of vulnerability assessment projects, that is, about one well per 3 km^2, during four seasons in 2008-09. In order to make any inferences about the areas that did not have well data, a statistical relationship between explanatory total organic carbon, electrical conductivity and nitrate variables related to well coordination was developed. The probability of the presence of elevated levels of the three compounds in the groundwater was predicted using the best-fit variogram model. According to spatial analysis, the highest R^2=0.789 achieved was related to electrical conductivity and followed the exponential model with 0.266 for NO_3^- (spherical model) and 0.322 for total organic carbon (exponential model) in the spring 2009. This showed the high confidence level for electrical conductivity dataset and forecasted trends. The results of the spatial analysis demonstrated that the transfer trends of electrical conductivity in the groundwater resources followed the route of groundwater movement in all seasons. However, for nitrate and total organic carbon, a definite trend was not obtained and pollution dispersion depended on many parameters.

Key words: Electrical conductivity, Nitrate, Total organic carbon, Variogram, Kriging

INTRODUCTION
One of the most important parameters for drinking and agricultural water quality is nitrate levels. Nitrate originates in surface and groundwaters from decomposed human and animal feces, industrial products, such as nitrogenous fertilizers and agricultural run-off (Di *et al.*, 2002). Generally, in precise planting, extreme use of fertilizer is controlled, due to the rising concentration of nitrogenous compounds in water drainage (Babiker *et al.*, 2004).
The annual application of nitrogen fertilizers and other crop management practices provide a considerable source of nitrates that may leach into groundwater in areas with at-risk soils and hydrogeology (Burkart *et al.*, 1999). Nitrate is the most frequently found chemical contaminant that is hazardous to human health in the world's groundwater aquifers (Gatseva and Argirova, 2008) and in many areas is becoming a serious worldwide environmental problem (Aslan and Türkman, 2005).
Nitrate concentrations have gradually increased in many European countries, such as the United Kingdom (an average annual increase of 0.7 mg/L), in the last few decades and have sometimes doubled over the past 20 years (WHO, 2007).
A study of nitrate levels in groundwaters of Iran, which was carried out in 2005, showed that the concentrations of nitrate ions in 42 water wells in Andimeshk and Shush plains (that cover an area of 1,100 km^2 between the Dez and Karkhe rivers in north of Khuzestan), had NO_3^-

*Corresponding author: E-mail: amin@hlth.mui.ac.ir
Tel: 0913167 42 54

concentrations (44.27 mg/L) below the United States Environmental Protection Agency (US-EPA) maximum contaminant level (MCL) and World Health Organization (WHO) guideline of 50 mg/L (Mahvi *et al.*, 2005).

Possible health concerns caused by nitrate ingestion are related to methemoglobinemia in infants less than 6 months of age after nitrate transformation in nitrite in the gullet, and to the possible formation of nitroso-compounds that are known to be carcinogens in the digestive system (Bouman *et al.*, 2002; Santafé-Moros *et al.*, 2005; Almasri and Kaluarachchi, 2007; Della Rocca *et al.*, 2007; Sadeq *et al.*, 2008). To protect against these effects, WHO and US-EPA have set the MCL for nitrate in drinking water at 50 mg/L (EPA, 2006; WHO, 2007).

Nitrate is mobile and permanent in many shallow groundwater environments (Takizawa, 2008). The increase of nitrate pollution in groundwater has led to the abandonment of numerous wells in agricultural zones (Lasserre *et al.*, 1999). An increase in concentration of this parameter can also imply that other contaminants, such as semi-volatile and volatile organic compounds (VOCs), metals, and inorganic pollutants, could be present in groundwater (Takizawa, 2008).

The polluting potential and strength of a given waste may also be estimated by measuring its carbon content. Because carbon reacts with oxygen, the more carbon the waste contains, the more polluting and stronger it is (Sincero, 2002). Total organic carbon (TOC) is a more precise and direct expression of total organic content. Measurement of TOC is of great significance to the operation of water and wastewater treatment plants and groundwater pollution sensing. Drinking water TOC ranges from less than 0.1 mg/L to more than 25 mg/L (APHA, 2005).

Today, in order to prevent the contamination of valuable freshwater resources, many countries and cities conduct groundwater vulnerability assessments as part of their urban development planning. Providing vulnerability maps enables cities to prevent future groundwater contamination and deterioration (Takizawa, 2008).

Electrical conductivity (EC) is a measure of the ability of the water to conduct an electrical current. EC of waters rise as different salts, depending on their type and quantity, are introduced into water resources (Qasim *et al.*, 2000).

Spatial models (Cressie, 1993) have been extensively used in many disciplines, such as groundwater pollution prediction to analyze dependent data collected from different spatial locations. Determination of the spatial correlation structure of the data and prediction are two important problems in statistical analysis of spatially distributed data. To solve them, a parametric variogram model is often fitted to the empirical variogram of the data. Since there is no closed form for the variogram parameters, they are usually compared numerically. The precision of spatial data analysis rigorously depends on properties of the random field, such as being Gaussian, stationarity and isotropicity. It also depends on how well a variogram model is fitted. Hence, a study of the data properties and fitting the best variogram model are the initial steps in an exploratory analysis of the spatial data (Iranpanah *et al.*, 2009).

From a usage point of view, the maps are considered to be fundamental tools for both national and local entities with appropriate responsibilities at the scheduling and management levels. At the consultancy level, applications appeared to be concerned mostly with the production of site-specific hazard and risk maps as an integral component of environmental impact studies (Mimi and Assi, 2009).

Considering the carcinogenic effects of some agrichemicals and regarding the fact that part of these chemicals may leach into groundwater, this study was designed in two parts. The first part provides basic groundwater quality information including EC as impurities quantity index, nitrate as inorganic and TOC as organic compounds index of industrial and agricultural activities in groundwater in the study area, and the second part included the spatial analysis of TOC, EC and nitrate (TEN) using GS^+ software. The results of this study should show pollution dispersion and prediction maps for groundwater quality in the area.

MATERIALS AND METHODS

Study area

Lenjan county with an area of 1093 km^2 is one of the cities which has been located 35 kilometers far from the city of Isfahan, in the southwest of Isfahan province and river plain of Zayandehrud. The barriers of this location are limited to Najafabad (north), the province of Chahar-Mahal and Bakhtiari (south and west), and Mobarakeh and Falavarjan cities (east). The city is divided into upper and lower parts (Olya and Sofla's Lenjan). The Sofla's Lenjan is placed in the east of Zayandehrud river, within the Najafabad plain aquifer. The land using in this area is mainly agriculture and is near to the industries such as acrylic textile, sugar beet and paper and cardboard.

Sampling wells

Among the 10,556 wells present in the study area, the estimated number of deep wells was 2,107, with average depths exceeding 50 m and with 323 Mm^3 discharge per year. Apart from that, there are 8,449 semi-deep wells with average depths of less than 50 m and a discharge rate of 507 Mm^3 per year and 73 subterranean wells with a discharge rate of 22 Mm^3 per year, and 1 spring with discharge rate of 1 Mm^3 per year. Approximate total intake of groundwater was reported as 853 Mm^3 in this area in 2005-2006.

From 1,730 km^2 flat surface in this area, approximately 80 km^2 of surface area was selected as the pilot in Sofla's Lenjan District (Najafabad, Isfahan, Central of Iran) that appeared to be most likely to be contaminated. By using the United States Geological Survey (USGS) guidelines, the groundwater sampling density in vulnerability assessment projects was about one well per 3 Km^2 (Johnson, 1998). Additionally, well selection was based on: (1) distribution of wells in the study area, (2) depth of the wells, (3) availability of well construction information, and (4) permission to access wells for sampling purpose. So, based on USGS guidelines, and on local considerations, as shown in Fig. 1, 25 deep wells were selected as sampling ports. When the derivation of the indicator is based on chemical analyses, the groundwater should be sampled quarterly, or at least twice a year (wet and dry periods) (Vrba and Lipponen, 2007). Thus, the sampling schedule was set at a quarter per year for the last two seasons in 2008 and the first two seasons in 2009.

Laboratory analysis

Groundwater samples were taken using well pumps after a pumping period of at least 30 minutes. Samples were transported to the laboratory in an ice bag and examined immediately for TEN according to standard methods (APHA, 2005).

All TOC measurements were performed using a Shimadzu-total organic carbon analyzer (Model: TOC-V_{CSH}, Japan), using high temperature combustion techniques (5310 B Method). The test method for nitrate determination was the 8039 method, the High Range Cadmium Reduction Method, using powder pillows in a range of 0.3 to 30.0 mg/L NO_3-N, in accordance with the Hach methods. All nitrate measurements were performed using a DR5000 UV/VIS Spectrophotometer (Hach-Lange, England). EC was determined on-site by a portable Hach Conductometer, Model 44600.

Spatial analysis and modeling

In this study, the kriging was used to spatially model the groundwater pollution based on laboratory analysis of nitrate, TOC and EC. The stationarity and isotropicity properties of a variogram are examined by spatial exploratory data analysis. Potential methods to fit a parametric variogram model of TEN have also been applied.

This study assumes that $Z(s_1),...,Z(s_n)$ are the realizations of a random field $\{Z(s), s \in D\}$, where $D \subset R^2$ and Z could be substituted with EC, TOC or nitrate, s is the sampling wells in different orientations, and D is the area under study. The spatial correlation configuration of the random field is determined by the variogram $2\gamma(s_1, s_2) = Var(Z(s_1) - Z(s_2))$ for all $s_1.s_2 \in D$. Under the intrinsic stationarity assumption, an unbiased estimator of the variogram is defined by:

$$2\hat{\gamma}(h) = \frac{1}{N(h)} \sum_{N(h)} [Z(s) - Z(s+h)]^2 \qquad (1)$$

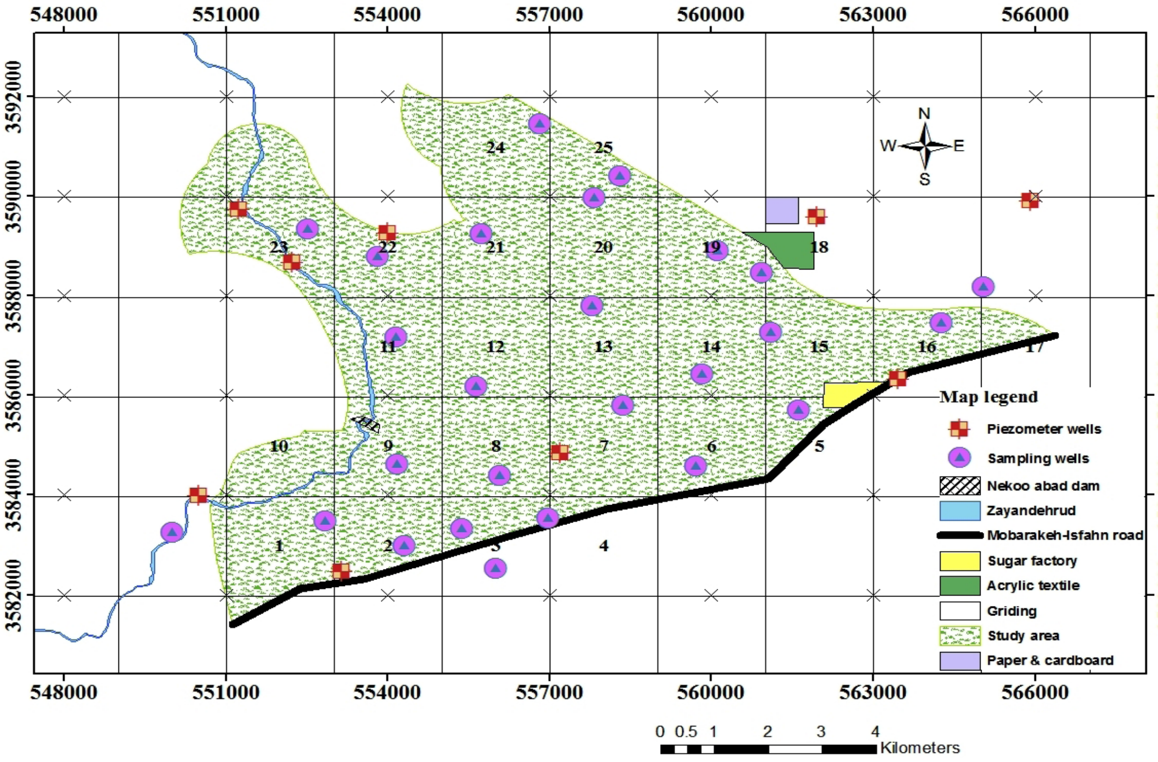


Fig. 1: Griding zone, piezometer and sampling wells in the pilot study area

Where:

$N(h)=\{(s_i,s_j):s_i-s_j=h;i,j=1,...,n\}$ and

N_h is the number of elements of $N(h)$. Since the variogram estimators (equation 1) cannot be used directly for spatial predictions, we needed to fit a valid variogram model that is closest to the spatial dependence structure present in the data. $P=\{2\gamma(.;\theta);\theta\epsilon\Theta\}$ denotes a parametric subset of valid variogram. To determine the best model that explains spatial structure of the dataset, several valid models are fitted to the empirical variogram. The two most exact parametric semi-variogram models are chosen:

(i) The exponential model:

$$\gamma(h:\theta)=\begin{cases}0, & \|h\|=0,\\ c_0+c_1\left(1-e^{-\frac{\|h\|}{a}}\right), & \|h\|\neq 0,\end{cases} \quad (2)$$

(ii) The spherical model:

$$\gamma(h:\theta)=\begin{cases}0, & \|h\|=0,\\ c_0+c_1\left(\frac{3}{2}\frac{\|h\|}{a}-\frac{1}{2}\left(\frac{\|h\|}{a}\right)^3\right), & 0<\|h\|\leq a,\\ c_0+c_{1,} & \|h\|\geq a,\end{cases} \quad (3)$$

Where $\theta=(c_0,c_1,a)$ consists of nugget effect, partial sill and range, respectively. Several methods, such as maximum likelihood (ML), restricted maximum likelihood (REML), ordinary least squares (OLS) and generalized least squares (GLS), can be applied to estimate θ (Iranpanah *et al.*, 2009). Considering the observations of $Z=(Z(s_1),...,Z(s_n))$, the ordinary kriging refers to the spatial prediction of the random field at a location s_0, under the assumption that $\mu(s) = E[Z(s)]$ is a fixed unknown real value μ, and the predictor is an unbiased linear function of the observations, i.e., $\hat{Z}(s_0) = \lambda' Z$ with coefficients $\lambda' = (\gamma - 1m)' \Gamma^{-1}$, where $m = -(1 - l'\Gamma^{-1}\gamma)/(l'\Gamma^{-1}l)$, $l = (1,\ldots,1)'$, $\gamma = (\gamma(s_0 - s_1),\ldots,\gamma(s_0 - s_n))'$ and Γ is the $n \times n$ matrix whose (i, j)th element is .The minimized mean-squared prediction error, namely the kriging variance, is given by $\sigma_k^2(s_0) = \lambda'\gamma + m$ (Cressie, 1993).

RESULTS

Table 1 shows the nitrate, EC and TOC concentrations of groundwater at 25 water wells that were sampled in this study. Fig. 2 shows spatial locations X (m) and Y (m) of EC from groundwater wells of the pilot study area in May 2009.

The quality of a fitted model to the spatial correlation structure of a dataset has an important effect on the precision of the predictors. The existence of several suitable parametric variogram models and different methods for the evaluation of their parameters give a spatial data analysis with many choices. Selection of the best model is essential for a complete study and to compare different models based on suitable criteria.

Fig. 3 illustrates an exponential semi-variogram model $\gamma(h;\theta)$ (equation 2) fitted to a classical semi-variogram estimate $\hat{\gamma}(h)$ (equation 1) with different distances h using GLS method for EC dataset.

Fig. 4 illustrates two dimensional (a) kriging surface $\{\hat{Z}(s_0) : s_0 \in D\}$ and (b) kriging standard-error surface $\{\sigma_k(s_0) : s_0 \in D\}$ for the best EC dataset with the variogram model $\gamma(h;\theta)$. Also, a two dimensional map is shown in Fig. 5 for EC dataset in May 2009.

Table1: Analytical results of TOC, EC and nitrate in groundwater wells

Area Code	Longitude (X)	Latitude (Y)	Nitrate ($mg/L\ NO_3^-$)				TOC (mg/L)				EC (mS/cm)			
	UTM, 39 N		Sept.08	Nov.08	Mar.08	May09	Sept.08	Nov.08	Mar.08	May09	Sept.08	Nov.08	Mar.08	May09
1	552826	3583474	13.2	--(1)	--	23.1	0.60	--	--	2.97	0.90	--	--	1.41
2	554300	3582979	72.7	--	28.9	47.6	21.6	--	1.84	1.20	4.25	--	4.48	4.12
3	555363	3583333	61.0	61.9	51.8	18.0	4.0	1.842	1.35	0.35	4.34	4.26	4.96	5.2
4	556970	3583541	44.0	43.4	25.0	42.3	2.13	1.441	1.58	1.61	--	4.65	4.58	3.82
5	561612	3585719	--	35.1	23.4	34.0	--	2.779	1.75	0.61	0.00	3.11	2.78	3.39
6	559707	3584593	29.0	20.2	--	26.7	0.70	0.855	--	0.76	2.06	2.32	--	2.13
7	558348	3585813	32.0	--	13.7	14.1	0.79	--	0.18	0.06	3.80	--	1.2	1.45
8	556066	3584396	15.2	20.8	21.8	19.1	1.60	1.179	3.56	1.11	0.95	1.06	1.03	1.08
9	554164	3584621	25.5	--	--	9.8	1.40	--	--	2.99	1.12	--	--	1.17
10	551575	3585220	11.5	11.9	6.6	--	0.62	2.437	0.77	--	0.73	0.88	0.82	--
11	554156	3587178	--	--	4.8	7.5	--	--	1.56	2.29	--	--	0.86	0.81
12	555797	3586985	49.2	28.5	17.6	27.1	--	0.3	3.21	2.01	0.96	1.06	1.03	1.24
13	557867	3587451	22.5	26.1	19.3	30.9	2.47	0.819	2.91	0.40	1.34	1.18	1.56	1.96
14	559464	3586950	23.1	13.0	--	21.3	1.24	0.427	--	0.63	1.52	1.8	--	1.8
15	561097	3587272	--	35.9	21.4	18.4	--	0.531	0.75	1.46	--	3.81	3.1	3.88
16	564256	3587469	18.9	21.1	15.4	16.7	--	1.382	4.04	2.25	4.49	6.81	4.99	5.5
17 (2)	--			--				--				--		
18	560929	3588479	25.7	41.5	17.1	17.1	1.10	0.53	1.82	1.93	7.10	2.78	2.97	8.63
19	560098	3588908	19.9	21.5	19.2	30.8	0.77	0.958	0.9	0.42	6.72	6.91	7.37	7.25
20	557814	3589973	23.9	--	18.5	22.0	1.16	--	0.80	1.69	4.88	--	3.4	6.31
21	556327	3589762	--	22.1	20.6	25.3	--	0.006	2.11	0.25	--	1.24	1.86	1.83
22	553815	3588793	20.2	--	14.2	10.9	2.28	--	3.47	0.12	0.82	--	1.32	1.1
23	552502	3589350	30.6	25.8	--	19.6	2.11	1.88	--	3.53	1.19	1.56	--	1.17
24	556819	3591457	22.9	16.2	14.5	25.0	0.28	0.288	0.13	1.70	1.42	1.23	1.25	1.75
25	558295	3590413	17.4	--	13.2	26.1	1.62	--	4.50	1.13	8.89	--	10.58	10.2
P & C (3)	561504	3590455	--	0.0	--	--	--	1.252	--	--	--	9.85	--	--
Zayandeh rud river	549546	3582351	5.3	5.3	6.1	11.9	--	--	2.69	4.73	--	1.24	1.15	2.12

(1) In the marked cells (--)due to lack of irrigation in this period of cultivation, wells had been shut down and there was not possibility for sampling.
(2) This zone has been located in a private district, so selecting a well and sampling was not implemented.
(3) P and C: the well was located in Paper and Cardboard factory.

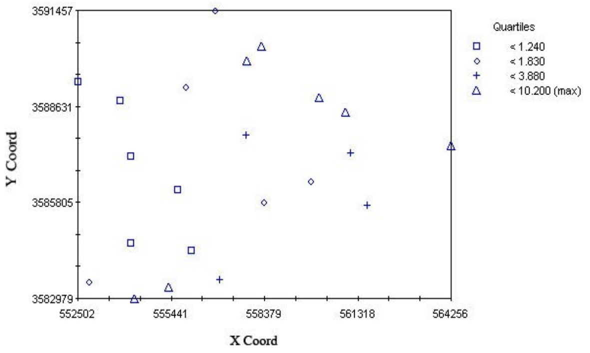


Fig. 2: Spatial locations x (m) and y (m) of EC in groundwater wells in May 2009

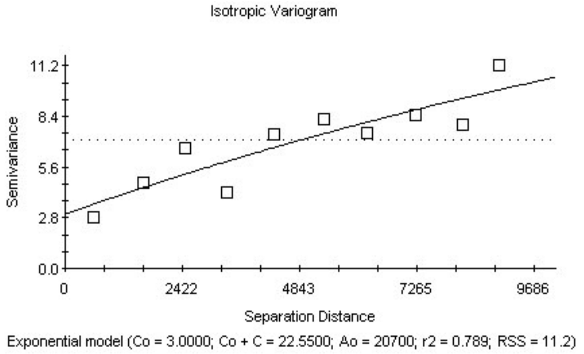


Fig. 3: Exponential semi-variogram model $\gamma(h;\theta)$ fitted to classical semi-variogram estimate $\hat{\gamma}(h)$ in different distances with GLS method for EC dataset in May 2009

As shown in the results, the spatial analysis was provided only for EC in the spring of 2009 as the best variogram model. These spatial analyses were also implemented for two parameters of TOC and nitrate in the study period; however, they are not illustrated here, except for kriging surface prediction maps which are shown.

Fig.6 shows kriging surface $\hat{Z}(s_0)$ for TOC, EC and nitrate dataset prediction for 2008-2009.

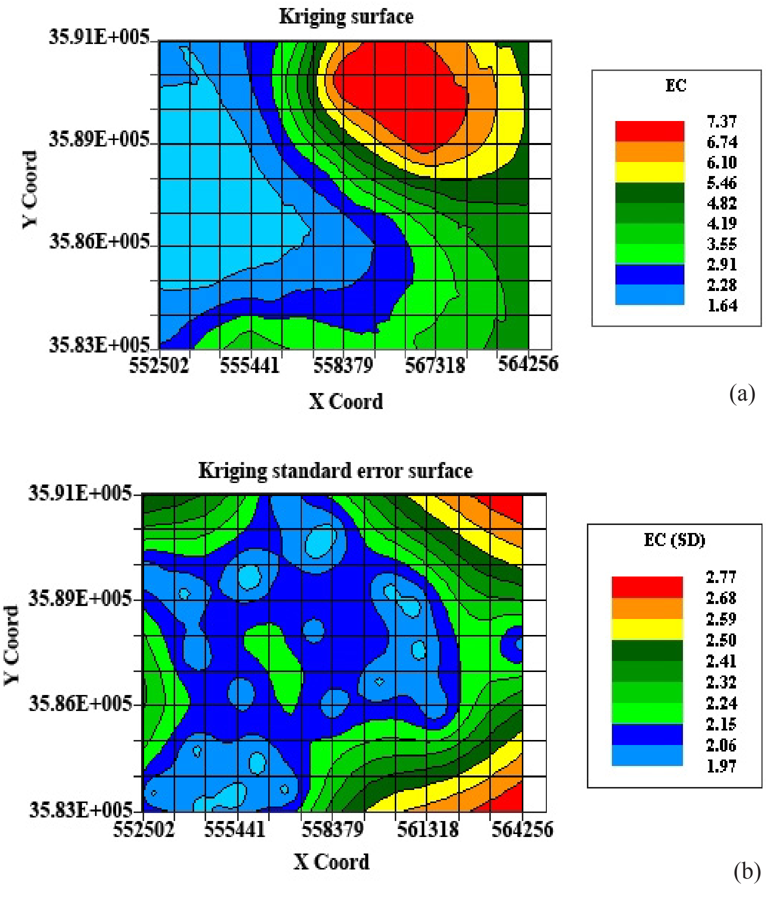


Fig. 4: (a) Kriging surface $\hat{Z}(s_0)$ and (b) kriging standard-error surface $\sigma_k(s_0)$ for EC dataset in May 2009

DISCUSSION

According to the EC semi-variogram in the spring 2009 in Fig. 3, any two locations with similar distance and direction from eachother have a similar difference squared. For the three parameters examined (TEN) in the spring of 2009, the highest R square coefficient (R^2=0.789) related to EC followed an exponential model, for NO_3^- was 0.266 (spherical model) and 0.322 for TOC (exponential model). The results showed a high confidence level for EC dataset and the forecasted trends. If a spatial correlation exists, pairs of points that are close together (on the far left of the x-axis) should have less of difference (be low on the y-axis). As points move farther away from each other (moving right on the x-axis), in general, the difference squared should be greater (moving up on the y-axis) (ESRI, 2009).

The kriging surface $\hat{Z}(s_0)$ of EC in Fig. 4.a indicates the trends of pollutant transfer in the spring season. The prediction map of this compound demonstrates that, as the distance from the river in the study area increased, the EC levels in the groundwater wells also increased. Thus, the downstream wells have higher salinity than those found upstream, causing problems for irrigation and potable water usage (Fig. 5). According to Fig. 4.b, that illustrates the kriging standard-error surface or confidence $\sigma_k(s_0)$ of EC levels, as the standard deviation of EC increases, the precision of the prediction map decreases. This may be due to the insufficiency of sampling wells in these regions.

As shown in the kriging surfaces of TEN in Fig. 6, the trend of EC-summer & autumn-08 and EC-spring-09 follow, the special pattern and is almost identical during the study period.

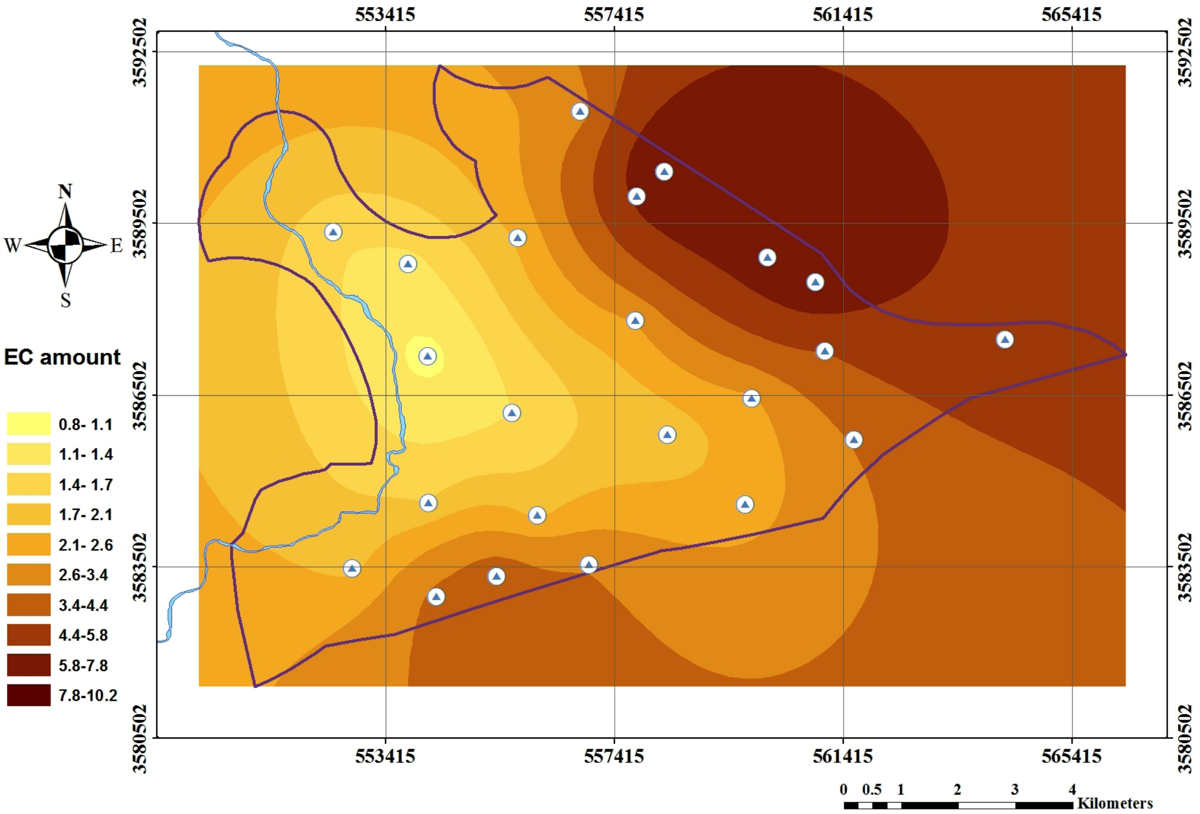
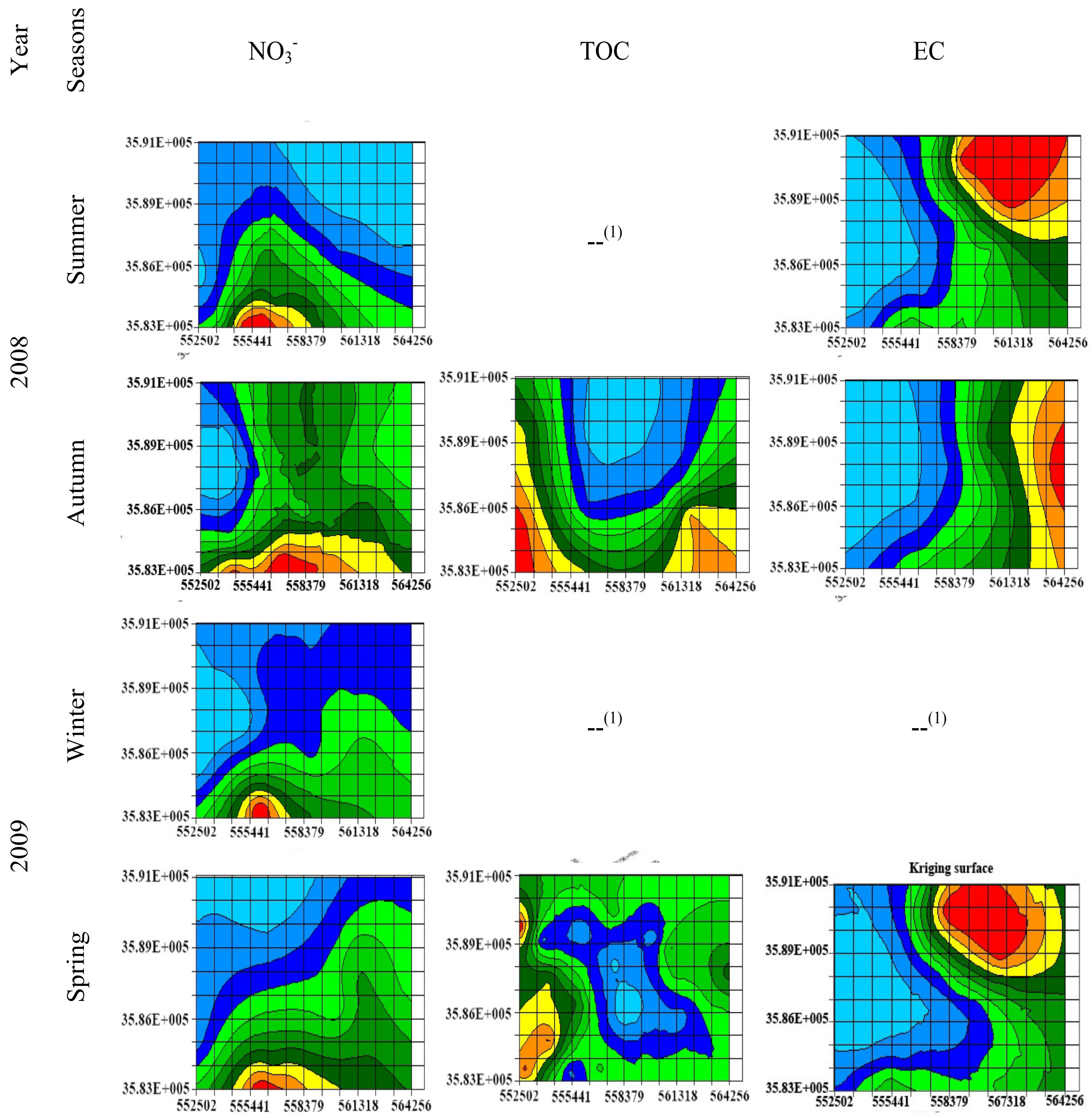


Fig. 5: Two dimensional map for EC dataset in May 2009



(1) In these seasons, the dataset has no spatial structure

Fig. 6: kriging surface $\hat{Z}(s_0)$ for EC, TOC and Nitrate dataset prediction in 2008-2009

EC is a measure of the dissolved ions in solution that come from the soils or bedrock through which the water travels, as well as from CO_2, which dissolves in precipitation as it falls to the ground (Brown *et al.*, 2007). EC generally increases along a groundwater flow path due to the combined effects of evaporation, ion exchange, and topographic conditions (Chen *et al.*, 2002; Brown *et al.*, 2007). Therefore if well drilling permitted downstream of the region, it is important to make farmers aware of the irrigation problems due to groundwater usage in this area. However, the kriging surfaces of TOC and nitrate in Fig. 6 show the nitrate during study period, TOC-autumn-08 and TOC-spring-09 have had abnormal trends. Therefore,

considering that the study area is one of the most important agricultural zones in the central region of Iran, the main cause of the fluctuations of NO_3^- and TOC could be the local heterogeneity of the groundwater flow pattern due to decreased water levels. This pattern could be induced by the farmers through the following factors:
1) the differences in pumping hours by the individual private wells for the seasonal planting of several crops that require various water consumptions; 2) the type and amount of chemicals, such as pesticides (measured as TOC) and nutritious compounds and fertilizers (detected as NO_3^-), that are used for the variant of crops; 3) the installation of new deep wells that lie in the influence radius of each other, which change the groundwater flow paths; and 4) geohydrochemical characteristics of the groundwater flowing towards the abstraction site.
According to some studies, the total area where an increment of concentration for a particular variable was detected means the sum of all areas where an increase in the concentration of nitrate or EC was detected during the observation period (Vrba and Lipponen, 2007). This is such that if the amount of water source EC is high, there exists the possibility that other pollutants, including concentrations of nitrate and other such compounds, are also high. Well depth was the dominant factor affecting NO_3^- concentrations (Spalding and Exner, 1993).
The results confirm an expected trend towards further degradation of the quality of the groundwater. The levels of nitrates are highest in the southern part, while the salinity affects primarily the wells in the northern part of the study region. Wells with deeper water levels that are located in northeast of the study area are less inclined to nitrate and TOC pollution and have excess salinity. In some wells, the NO_3^- concentrations increased significantly in groundwater due to leaching. The frequency of wells with nitrate levels that exceed 50 mg/L is higher in residential areas, such as zones 1, 2 and 3 in Figure 1 that have a higher population density than in the other zones. This suggests that inadequate sanitation may enhance the risk of nitrate pollution. Since some of the agricultural wells in the study area are also used as drinking water sources, monitoring the groundwater quality is extremely important.
Different methods for assessing groundwater vulnerability have been presented around the world. In this study, a spatial analysis method was used to predict the movement of three pollutants (TEN) in groundwater of the project area. Therefore, approximately 80 km^2 representing the total area of 1,730 km^2 was selected as the pilot study area in the central plateau of Iran.
The results of the spatial analysis showed the EC transfer trends in the groundwater resources, which were due to factors like the length of travel of the water and other parameters could follow the route of groundwater movement all year long. Whereas it is necessary to dig a well in the downstream area of the region (northeastern region of study area), this should be further investigated to ensure that there is no possibility of contamination of these resources. However, for nitrate and TOC, there is no specific trend and the distribution of these pollutants depends on water withdrawal and the consumption of compounds, including pesticides and fertilizers in different areas.
It is recommended that a spatial analysis of the entire catchments in the Najafabad aquifer to be undertaken. This should be accomplished with increasing the number of water quality monitoring stations. An application of method used herein requires the continuous sampling of water resources. Therefore, in order to achieve the desired results, utilizing software such as the Geographical Information System (GIS), is helpful. GIS could be used online to monitor water resources over a longer time and using satellite images to obtain trends in the spatial and temporal changes of the total groundwater quality parameter distributions.

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