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Original article

Spatial prediction of chloride concentration in Azarshahr plain aquifer-Iran, using EC, Ca^{++} and Mg^{++} as auxiliary co-kriging variables

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A B S T R A C T

For a less densely sampled Area, Lognormal ordinary cokriging (LnOCK) with auxiliary variables can sometimes improve estimates. In this study for groundwater quality assessment of Azarshahr plain aquifer- East Azerbaijan province- Iran (one of the Uromia lake sub-basins) 39 samples have been gathered. Due to slight sample accumulation, geostatistics was utilized for accuracy rising in Chloride concentration prediction of study area. For this purpose, three steps were designed; At first, spatial concentration of chloride has modelled by Lognormal ordinary kriging (LnOK), then three different covariant (EC, Ca^{++} and Mg^{++}) that their quantities had more than 90% correlation to chloride, has been chosen for spatial prediction of its concentration separately and in third step EC and Ca^{++} have used together as covariates to evaluate the spatial prediction. Outcomes have shown that Lognormal ordinary cokriging (LnOCK) using Ca^{++} as an auxiliary covariant reveals more efficient results; With Mean error about 0.04 and RMSE about 0.26. Whereas adding more data set as excess covariant (Ca^{++} and EC, together) reduced the model precision. Drown maps finally showed that Chloride concentration rises from the South-East to North-West in study area.

1. Introduction

Nowadays, groundwater is a major source of supply for domestic and agricultural purposes; especially in arid and semi-arid regions. Groundwater systems possess features such as complexity, nonlinearity, being multi-scale and random, all governed by natural and/or anthropogenic factors, which complicate the dynamic predictions (Nourani, 2012). Huge quantities of groundwater, particularly from the shallow aquifers, are used for potable use and irrigation in Iran. In the absence of adequate surface water in the dry season; irrigation becomes heavily dependent on groundwater (Moasheri et al., 2012). Understanding the behavior of the groundwater body and its long term trends are essential for making any management decision in a given watershed (Reghunath et al., 2005). Therefore, having a deep knowledge and insight on the groundwater system seems necessary for optimum exploitation of water. It is now recognized that quality of groundwater is just as important as quantity (Todd & Mays, 2005).

Pollution of groundwater recently emerged as a globally growing environmental problem due to the increasing demands of groundwater for drinking and agricultural purposes. In the other hand groundwater can affects water quality in many regions because of its salinity. In many management works, it is necessary to know the spatial and temporal behavior of groundwater. Water quality measured according to some parameters that chloride is one of them, so it is important to measure chloride in irrigation and potable water for suitable management and yield maximization (Rostami Zad et al., 2011). Chloride occurs in all natural water in widely varying concentration. The chloride content normally increases as the mineral content increases. Upland and mountain supplies usually are quite low in chloride, whereas rivers and groundwater usually have considerable amount (Sameer et al., 2010). Chloride in groundwater derives from both natural and anthropogenic sources, such as runoff containing road deicing salts, the use of inorganic fertilizers, landfill leachates, septic tank effluents, animal feeds, industrial effluents, irrigation drainage, and seawater intrusion in coastal areas (Sameer et al., 2010).

The variations of groundwater quality over years in many parts of Iran, suggest a precise and detailed study to be undertaken to elucidate the behavior of groundwater chemistry fluctuations. A very useful tool for analyzing such processes is geostatistic (Ahmadi and Sedghamiz, 2007). Geostatistic refers to the sub-branch of spatial statistics in which the data consist of a finite sample of measured values relating to an underlying spatially continuous phenomenon. Examples include: heights above sea-level in a topographical survey; pollution measurements from a finite network of monitoring stations; determinations of soil properties from core samples and etc. Originally, the term geostatistic was coined by Georges Matheron and colleagues at Fontainebleau, France, to describe their work addressing problems of spatial prediction arising in the mining industry (Diggle and Ribeiro Jr, 2007).

In these years geostatistic plays an important role in distributed models and spatial analyzing for instant Voudouris et al. (2004) described methods for defining the areal salinity distribution by seawater intrusion by geostatistic; they collected and analyzed samples from two representative aquifers of Greece, T.D.S., Cl^{-} concentration, Br^{-} concentration, and analysis of the salinity factor and hydrochemical sections had done. They computed Experimental and theoretical semivariograms of the selected parameters. Maps had shown geographical distribution, using the ordinary kriging method. From these maps, the seawater intrusion zone had defined. Lei et al. (2008) had employed multivariate statistical and geostatistical methods to identify spatial variability of trace elements in Agricultural Soils in Dongguan City, Guangdong, China; they collected samples from agricultural fields, including vegetable and orchard soils in the city, and analyzed eight heavy metals (As, Cu, Cd, Cr, Hg, Ni, Pb, and Zn) and other items (pH values and organic matter) , to evaluate the influence of anthropic activities on the environmental quality of agricultural soils and to identify the spatial distribution of trace elements and possible sources of trace elements, results had shown that the source of pollution derived from three origin (natural source, industrial and traffic pollution sources and long-term anthropic activities). Hani (2010), tested the concentrations of As, Hg, Co, Cr and Cd for soil samples in Kaveh industrial city, and analyzed their spatial patterns by the semivariogram approach of geostatistic and geographical information system technology. Results of study was helpful for risk assessment of environmental pollution for decision making for industrial adjustment and remedy soil pollution. Ghadermazi et al. (2011) put the aim of their study on comparison lognormal ordinary cokriging (LnOCK) with lognormal ordinary kriging (LnOK) and lognormal inverse distance weighting (LnIDW) for the spatial prediction of NO3-N in drinking water using pH as an auxiliary variable in LnOCK their study had revealed that In terms of mean error (ME) and root mean squared error (RMSE) LnIDW performed much better than LnOK for NO3-N. However, LnIDW was consistently less effective than LnOCK using pH as auxiliary variable.

2. Materials and methods

2.1. Study area

The Azarshahr Plain is one of sub-basins of the Uromia Lake watershed, is located in Azarbaijan province, northwest of Iran. The plain is bordered to the east and southeast by volcanic Sahand Mountain and to the south by travertine of Ghezeldagh, to the north and west by Aji Chay and Uromia Lake salty flat plain respectively (fig.1). The study area is a densely populated area of Iran, with 100 percent of its drinking, domestic and industrial water and 80 percent of agricultural water supplied from groundwater resources (Asghari Moghaddam, 1991). The total area of the Azarshahr basin and Plain are about 580 km2 and 136 km2 respectively.

According to Azarbaijan Regional Water Authority (ARWA) (2009), the Hydrological and meteorological properties of study area, are as follow: Average annual precipitation values is about 221/2 mm for a 30 years period; whereas its mean evaporation is about 1490mm, shows the important role of groundwater in the study area. Mean daily temperatures vary from 0.14ºC in January up to 25.8ºC in July with a yearly average of 13º C and.

Azarshahr Chay is the main river in the study area, which is originated from Sahand Mountain (east of the area) and it inters the plain from east side and through the plain discharges in to the Uromia Lake. The river rarely discharges into the Lake due to percolation and evaporation losses, as well as diversion of water for irrigations.

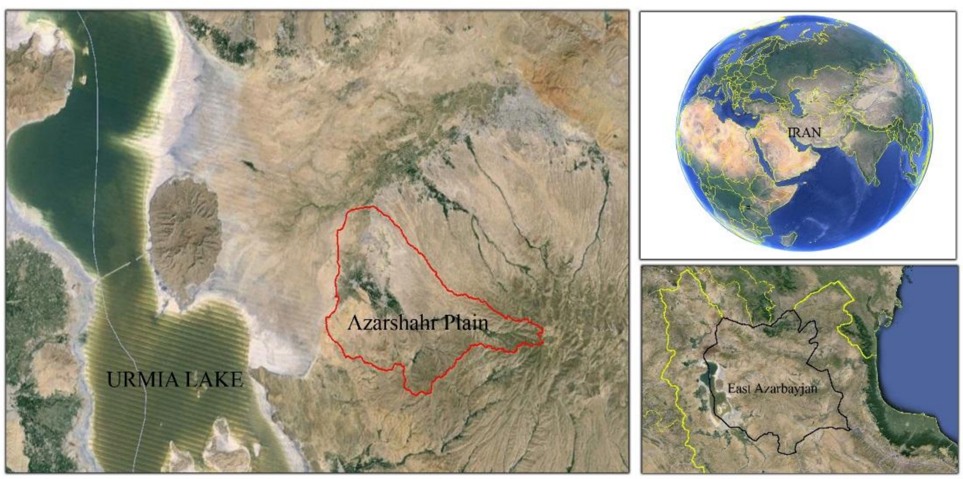


Fig. 1. Geographical location of the study area.

2.2. Geological setting

Due to geology effect importance on groundwater quality, precise insight is necessary about the geology of study area. The study area lies in East Azarbaijan province, which is structurally part of Central Iran unit. It is wedged between the Zagros and Alborz mountain systems. The area includes representatives of Jurassic to Quaternary age with various movements affecting it, most strongly those of Alpine origin. Pliocene time involved a marine regression and a change to continental conditions, mainly lacustrine, coupled with the deposition of clay and clastic. Then the Plio-Pleistocene was marked by significant volcanic activity, with lava flows and pyroclastic masses associated with the continental conditions of that epoch.

Hence, the eastern part of the Azarshahr area is occupied by the extinct Sahand volcano, which is built up from a volcanic series of rocks. This massif is surrounded by volcanic sediments called “alluvial tuff”, which were deposited around the andesitic core (Moinvaziri et al., 1975). The Sahand alluvial tuff conformably overlies Pliocene marls, sandstones and fish-bed layers (the bedrock of the study area). The southwestern part of study area includes Jurassic and Cretaceous limestone with Pliocene travertine, which is believed to be connected to the thermal mineral issuing from the Cretaceous limestone as well as from alluvial tuff (Issar, 1969).

The alluvial water course and plain deposits of the study area are derived from the erosion of Sahand pyroclastic materials, which have transported by water and other transporting agents and deposited in the Azarshahr Plain. They are coarse and poorly sorted in the highest parts of the plain and become progressively finer and more clayey towards the Uromia Lake, which is flanked by a salty loam and huge clay plug. From geological point of view the Quaternary alluvial deposits including water course and plain deposits forming the main water bearing layers in the study area. Some of Qanats and springs are originated from alluvial tuffs of Sahand Mountain and come up from the boundary of these formations and Pliocene marls and fish-beds. Figure 2 depicts a schematic geological view of study area.

The alluvial aquifer of the study area has been known for many years as a good aquifer, through Qanats, geophysics and well distributed drilled wells. It has been extensively developed for public and agriculture water supply and investigated hydrogeologically, particularly in connection with groundwater development. According to Azarbaijan Regional Water Authority (ARWA) (2009), 233 deep and 500 shallow active pumping wells, 162 Qanats and 6 springs operate in the alluvial aquifer of the plain that imposes stress to the aquifer quantity and quality of course.

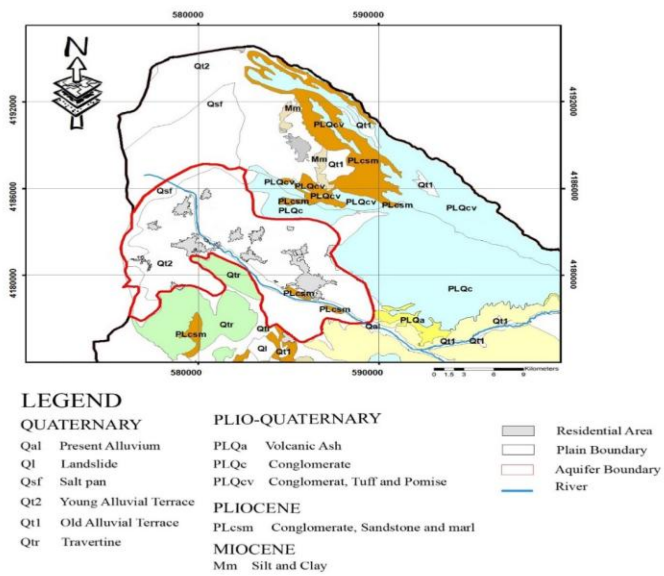


Fig. 2. Geological setting schematic view of study area.

2.3. Theoretical basis

The study of groundwater chemistry provides important clues on the geological history of the water bearing layers, gives some indication of groundwater recharge, and the velocity and direction of flow patterns and storage (Freeze and Cherry, 1979); but it is not always possible to examine every location quality. Therefore, unknown values must be estimated from data taken at specific locations that can be sampled. The size, shape, orientation, and spatial arrangement of the sample locations are termed the support and influence the capability to predict the unknown samples. For Azarshahr study area, gathered samples did not cover all the study area.

A unique aspect of geostatistics is the use of regionalized variables which fall between random variables and completely deterministic variables. Regionalized variables describe phenomena with geographical distribution (e.g. elevation of ground surface).This phenomenon exhibit spatial continuity

The theoretical basis of geostatistics has been fully described by several authors (Chiles, 2012; Diggle, 2007; Goovaerts, 1997; Hengl, 2009; Isaaks and Srivastava, 1989; Kitanidis, 1997 and Webster and Oliver, 2001).

Regionalized variable theory uses a related property called the semivariance to express the degree of relationship between points on a surface. The semivariance is simply half the variance of the differences between all possible points spaced a constant distance apart. Semivariance is a measure of the degree of spatial dependence between samples (here Chloride concentration) .

The Semi-variogram, γ (h), can be defined as one half the variance of the differences between the attribute values at all points separated by a distance h as follows:

$$\gamma(h) = \frac{1}{2N\,(\mathrm{h})}\sum_{i=1}^{N\,(\mathrm{h})}[Z\,(\mathrm{X}_i) - \mathrm{Z}(\mathrm{X}_i + h)]^2 \qquad (1)$$

Where Z(x) indicates the magnitude of variable, and N (h) is the total number of pairs of attributes that are separated by a distance h Figure 3 shows the Semivariogram of the Chloride concentration.

Prior to the geostatistical estimation, we require a model that enables us to compute a variogram value for any possible sampling interval. The most commonly used models are Spherical, Exponential, Gaussian, and Pure nugget effect (Isaaks and Srivastava, 1989) that in our case Exponential model was the best fitting curve for samples. The adequacy and validity of the developed variogram model was tested satisfactorily by a technique called crossvalidation. The idea of cross-validation consists of removing a datum at a time from the data set and re-estimating this value from remaining data using different variogram models (Rostami Zad et al., 2011).

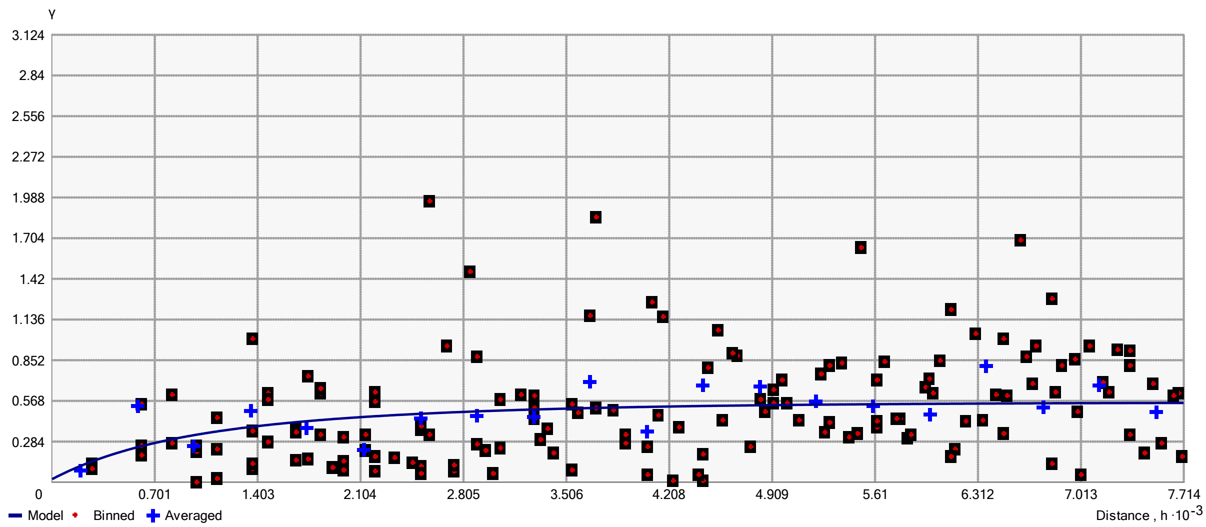


Fig. 3. Semivariogram of Chloride concentration.

After model selection we must start the interpolation and prediction process subsequently using Kriging interpolation and prediction method in this study. Kriging is given to a class of statistical techniques for optimal spatial prediction. Kriging technique is a spatial interpolation estimator Z (X_0) used to find the best linear unbiased estimator of a second-order stationary random field with an unknown constant mean:

$$Z\,(\mathrm{X}_0) = \sum_{i=1}^{n}\lambda_i Z\,(\mathrm{X}_i) \qquad (2)$$

Where Z (X_0) is Kriging estimate at location X_0; Z (X_i) is sampled value at X_i ; λ_i is weighting factor for Z (X_i); and i = 1… n in which n denotes to the numbers of samples.

Kriging uses the semivariogram, in calculating estimates of the surface at the grid nodes. In the kriging method, every known data value and every missing data value has an associated variance.

Based on the semivariogram used, optimal weights are assigned to known values in order to calculate unknown ones. Since the variogram changes with distance, the weights depend on the known sample distribution.

Ordinary kriging is the simplest form of kriging; it uses dimensionless points to estimate other dimensionless points, e.g. elevation contour plots. In Ordinary kriging, the regionalized variable is assumed to be stationary. In our case Z, at point p, Z_e (p) to be calculated using a weighted average of the known values or control points:

$$Z_e(\mathrm{p}) = \sum W_i \times Z\,(\mathrm{p}_i) \qquad (3)$$

Figure 4 shows the map of Chloride distribiution using Ordinary kriging and its crossvalidation graph.

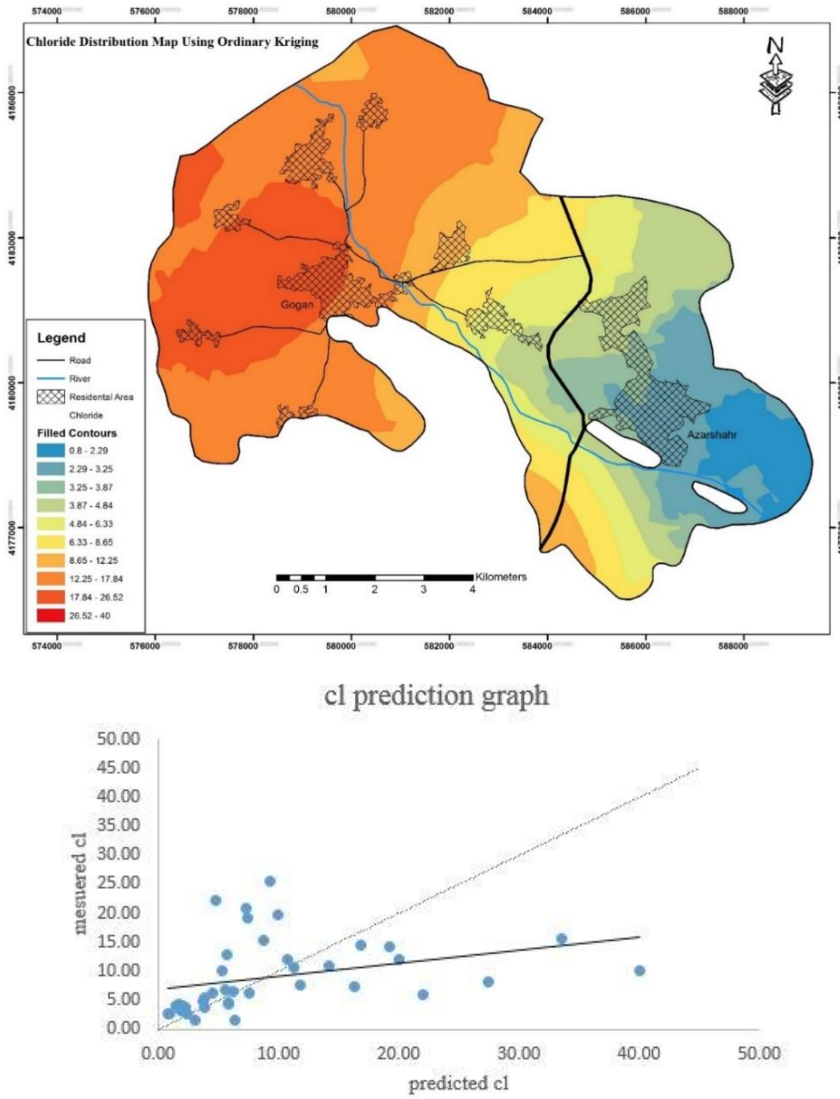
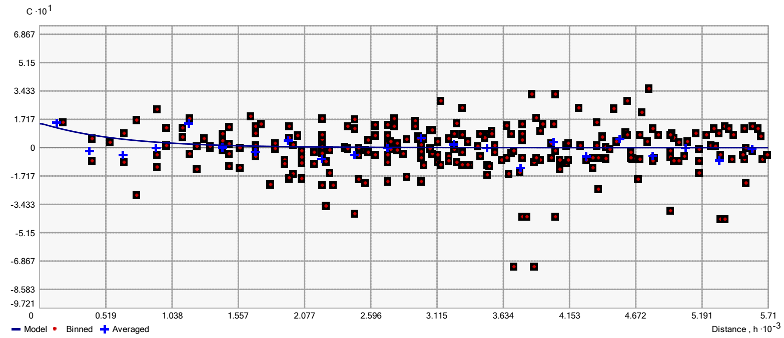
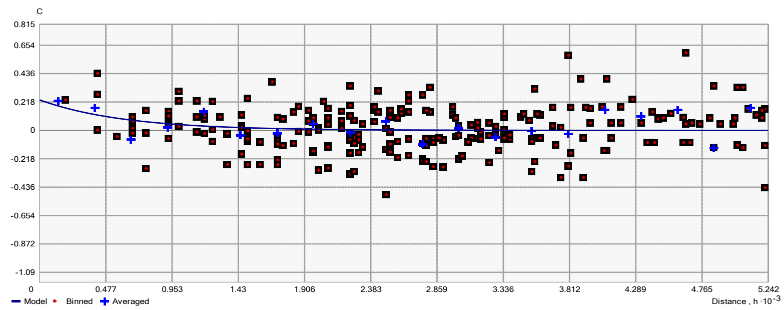


Fig. 4. Chloride prediction map using Log normal ordinary Kriging and crosscvalidation graph.

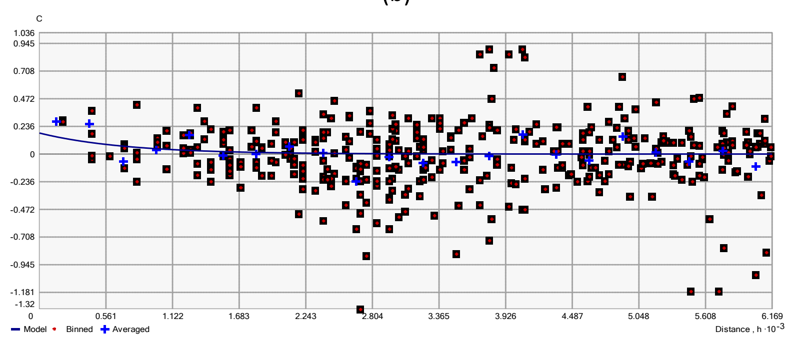
Kriging using information from one or more correlated secondary variables, or multivariate kriging in general. The "co-regionalization" (expressed as correlation) between two variables, i.e. the variable of interest, groundwater chloride in this case and another easily obtained and inexpensive variable, can be exploited to advantage for estimation purposes by the Cokriging technique (Nourani, 2012). Figure 5(a, b, c and d) shows the crosscovariance of Chloride with Calcium, Electrical Conductivity, Magnesium and Calcium and Electrical Conductivity together as covariant respectively.



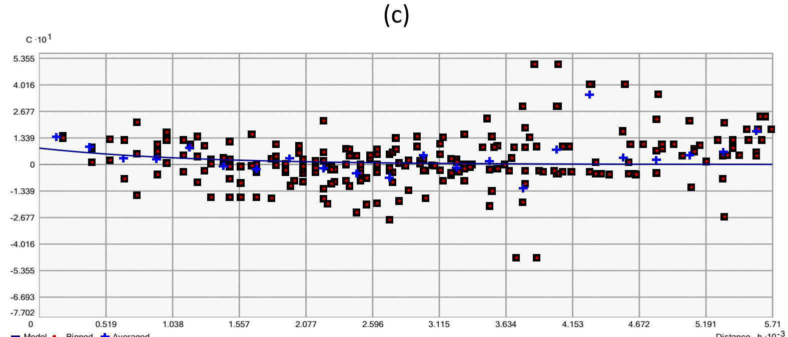
(a)



(b)



(c)



(d)

Fig. 5. Co-variance of Chloride with Ca^{++}, EC, Mg^{++} and Ca^{++} and EC together

3. Results and discussion

39 sample have been collected and analyzed for major elements and also nitrate, fluoride and some trace elements from study area. Figure 6 depicts the sampling points in the study area; hydrochemical evaluation revealed that Cl^{-} and Ca^{++} frequency are more than other elements (Table 1), then The Piper diagram mapped for samples (Figure7). After that Chloride due to its importance had chosen for distributed modelling; some

statistical parameters have derived for chloride (Figure8). For Chloride spatial prediction, geostatistical Analyst module have used from Arc map GIS software. At first, Log normal Ordinary Kriging have used, the map of prediction and computed crossvalidation has drown. In the second step of study correlation between Cl^{-} and other parameter such as EC, PH and some elements such as Ca^{++}, Mg^{++}, K^{+} and Na^{+} evaluated and revealed that EC and Ca^{++} have more correlation than the others (Figure 9).

Table 1
Sample Major Elements Concentration.

Sample	X	Y	So4(mg/l)	Cl(mg/l)	Hco3(mg/l)	Ca(mg/l)	Mg(mg/l)	Na(mg/l)
1	582896	4192518	330.12	324.90	256.20	288.72	29.16	84.64
2	583631	4191801	41.74	299.91	209.84	118.70	38.77	77.42
3	587076	4185730	106.86	128.96	158.60	107.47	16.52	58.15
4	586803	4188658	85.93	92.97	136.64	75.39	14.58	55.74
5	585453	4189431	77.79	64.98	195.20	97.84	1.94	50.92
6	583960	4189666	464.53	969.70	246.44	481.30	145.80	111.14
7	583531	4187539	698.44	1139.65	251.32	425.06	111.78	482.42
8	582834	4183303	508.07	209.93	373.32	232.58	34.02	173.76
9	583113	4183390	511.05	284.91	512.40	258.24	54.13	209.90
10	582150	4184456	509.56	299.91	366.00	279.10	47.33	159.31
11	581536	4183402	563.37	464.86	561.20	312.88	106.92	226.13
12	580635	4182186	170.81	199.94	231.80	210.12	9.72	79.83
13	579854	4184804	301.74	1079.67	390.40	497.24	140.94	159.55
14	580796	4184738	1278.49	1519.53	341.60	938.34	165.24	260.48
15	581508	4184819	1621.51	684.79	222.04	565.56	145.80	376.43
16	582397	4181364	206.86	224.93	536.80	245.41	15.14	128.00
17	581415	4181233	342.44	249.92	517.28	288.82	29.16	115.96
18	579762	4182524	301.74	1329.59	512.40	681.70	106.92	210.13
19	578038	4183214	325.00	1519.53	422.12	721.80	131.22	228.08
20	577687	4181034	604.07	2249.30	573.40	882.20	199.26	381.25
21	577796	4181909	447.09	1149.64	673.44	617.54	160.38	193.03
22	577141	4179695	604.07	479.85	519.72	449.12	48.60	173.76
23	576440	4181071	1115.70	3039.06	444.08	1154.88	456.84	704.69
24	578889	4179896	336.63	1084.66	727.12	425.06	140.94	366.80
25	578293	4179993	252.26	894.72	722.24	537.34	68.04	226.76
26	580074	4180296	295.93	239.93	1073.60	352.88	97.20	168.95
27	580377	4179377	394.77	544.83	1098.00	309.22	126.36	357.16
28	582656	4180267	240.58	209.93	456.28	180.60	47.33	131.73
29	583926	4179404	137.09	104.97	463.60	144.36	34.02	62.96
30	587685	4177975	68.49	38.98	329.40	101.05	14.58	31.65
31	584283	4181119	156.53	209.93	439.20	179.65	36.94	96.69
32	584510	4178599	110.35	94.97	397.72	117.09	30.13	60.56
33	583281	4176378	1092.30	539.83	1427.40	573.44	140.94	492.05
34	585718	4177555	140.58	99.97	459.33	121.90	23.33	106.32
35	588862	4179008	71.68	34.98	368.44	85.01	12.64	60.56
36	586990	4182722	108.02	154.94	207.40	96.24	37.91	55.74
37	584355	4182095	119.65	144.94	234.24	97.84	34.02	55.74
38	584953	4182683	166.35	244.92	373.32	187.67	30.77	94.28
39	582357	4185585	97.56	1839.47	387.96	673.68	213.84	202.67

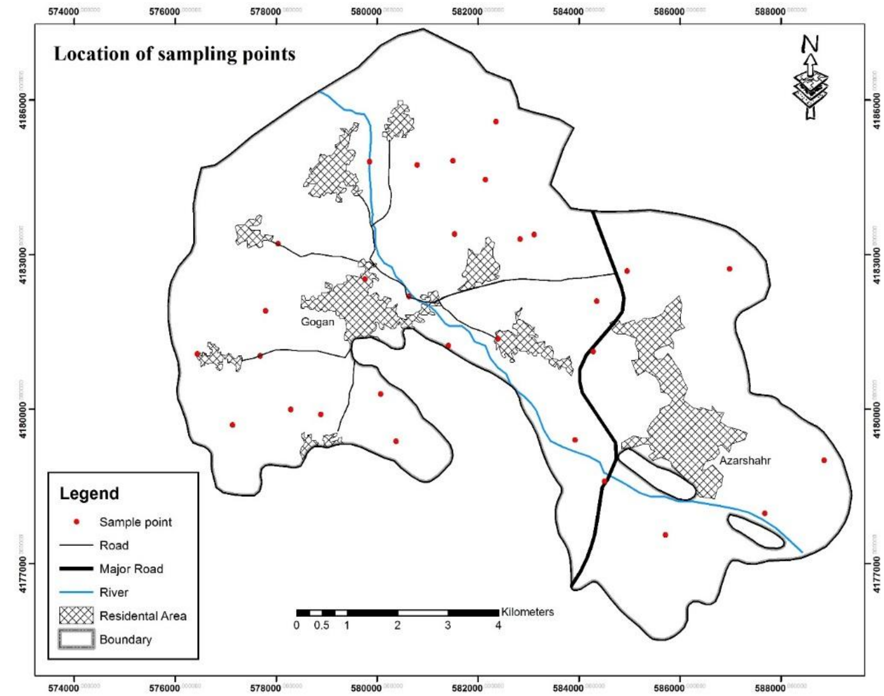


Fig. 6. Sampling point in study area.

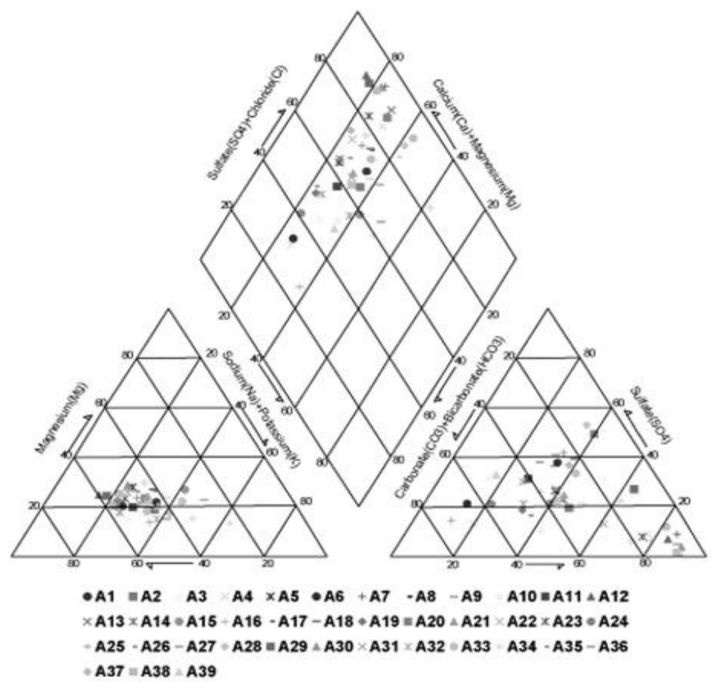
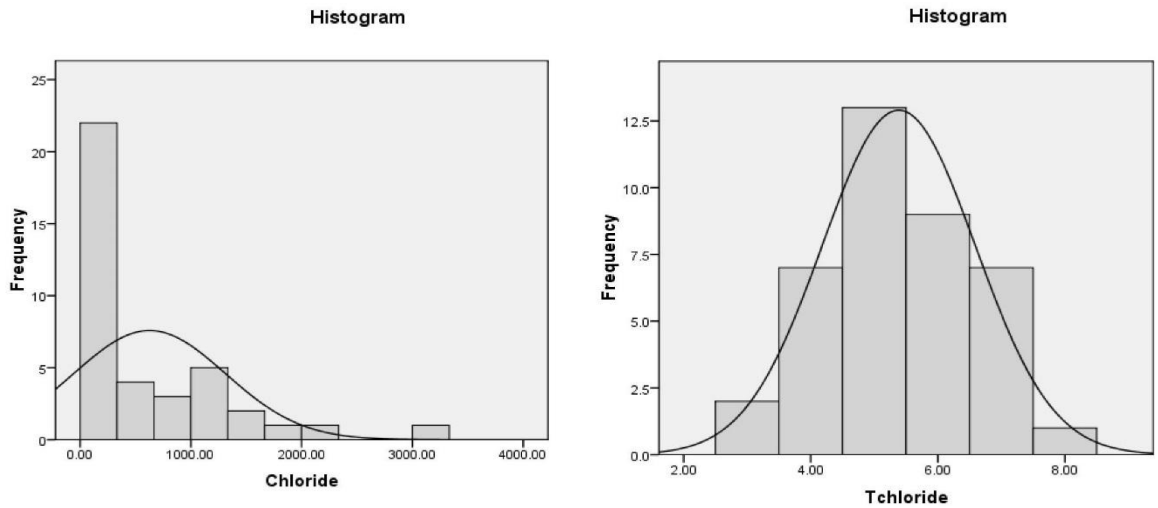


Fig. 7. Piper Diagram for samples.

The commonly used descriptive parameters were calculated (Table 2 and 3), Histograms of Chloride density with a normal distribution curve are shown in Fig. 8. The raw data have a long tail towards higher Chloride density values (Fig. 8a). Other studies have also shown that environmental variables are often skewed from a normal distribution towards positive values because of the relatively smaller percentage of high values (Chang et al., 2003). The Ln transformed data show a normal distribution (Fig. 8b). This is confirmed by the K–S p value (> 0.05). Therefore, transformed data were used for geostatistical analysis.



(a) (b) **Fig. 8.** Histogram of Sample Chloride Concentrations.

Table 2
Descriptive Parameters of Chloride before Normalization

Min	Mean	Max	Skewness	Kurtosis	Kolmogorov-Smirnov	Shapiro-Wilk
34.98	627.10	3039.058	1.746	3.188	0.000	0.000

Table 3
Descriptive Parameters of Chloride after Normalization.

Min	Mean	Max	Skewness	Kurtosis	Kolmogorov-Smirnov	Shapiro-Wilk
1.01	5.38	13	0.046	0.729	0.2	0.498

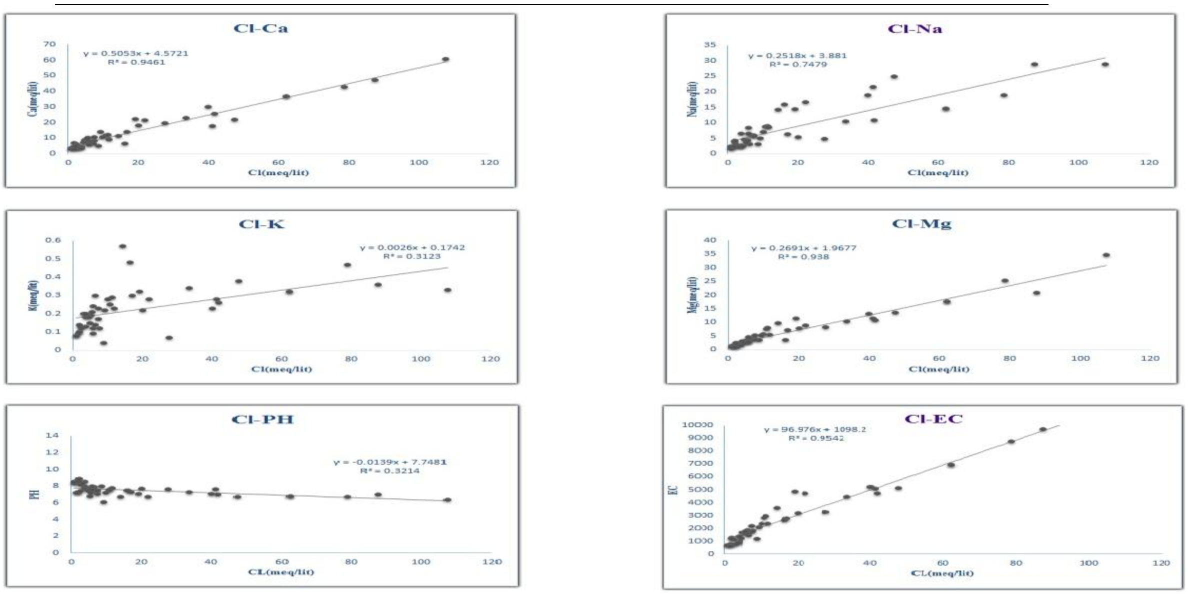


Fig. 9. Correlation between Cl^{-} and other chosen parameters and elements.

As Figure 9 shows, the correlation between Cl^{-} and EC and its correlation with Ca^{++} is 0.9542 and 0.9461 respectively. Also Mg^{++} has a correlation about 0.94. So these 3 cases were chosen as covariant in the prediction of Cl^{-} distribution with lognormal ordinary cokriging (LnOCK). Whereas Na^{+} correlated about 0.747 with Cl^{-}. Upon to Hounslow (1995), if chloride be more than Sodium, then there is an analytical error or the composition of the water is derived from brines where reverse ion exchange or reverse natural softening has occurred. In the latter case, one would expect the dissolved solid content of the water to be high- at least over 500 Mg/lit. Also if Ca be more than So4 indicates Ca^{++} source other than gypsum, such as Calcite/Dolomite or silicates (Figure 10). Considering the geology of study area reveals that Ca^{++} sources is mainly derived from Travertine member. Figure 11 also shows the distribution of Ca^{++} in the study area.

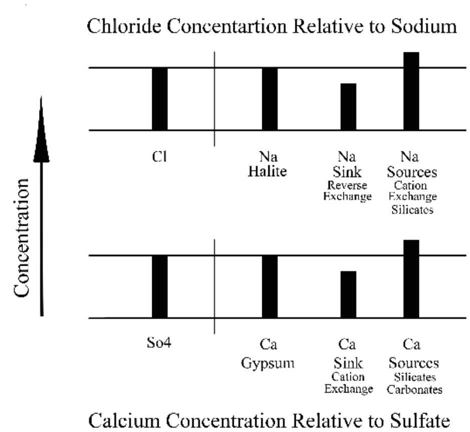


Fig. 10. Origin of Ca and Na considering the Cl concentration (Hounslow. 1995).

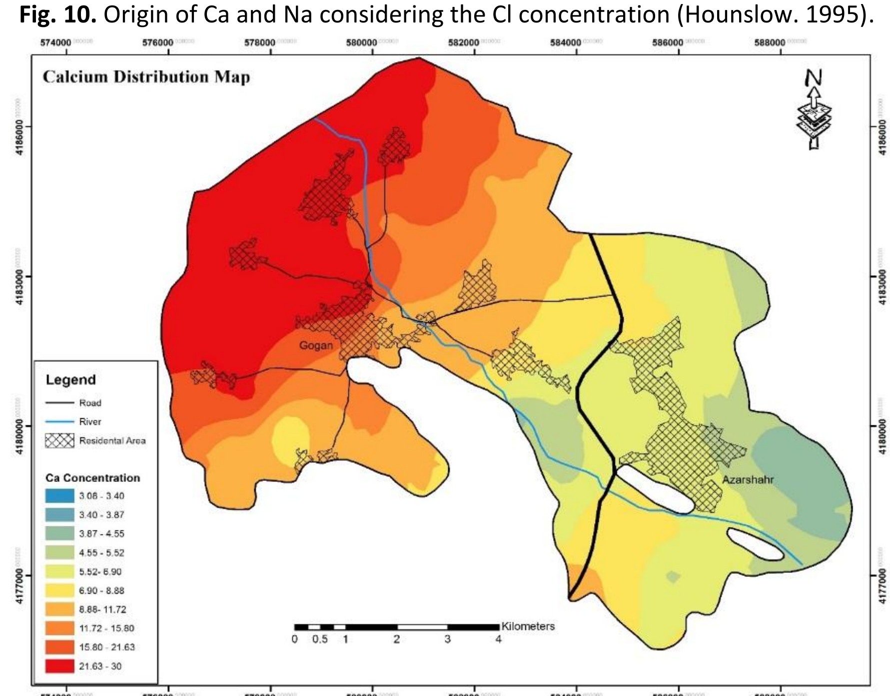


Fig. 11. Distribution of Ca^{++} in the study area.

After using cokriging method for prediction, the results figure out in the study area map (Figures 12 to 14) and then their crossvalidation have computed (Figure 15). In third step Ca^{++} and EC have used together as covariates in lognormal ordinary cokriging (LnOCK), to increase the prediction of Cl^{-} distribution; result has shown that conjugating of these covariates had no positive effect on prediction (Figure 16, Table 4).

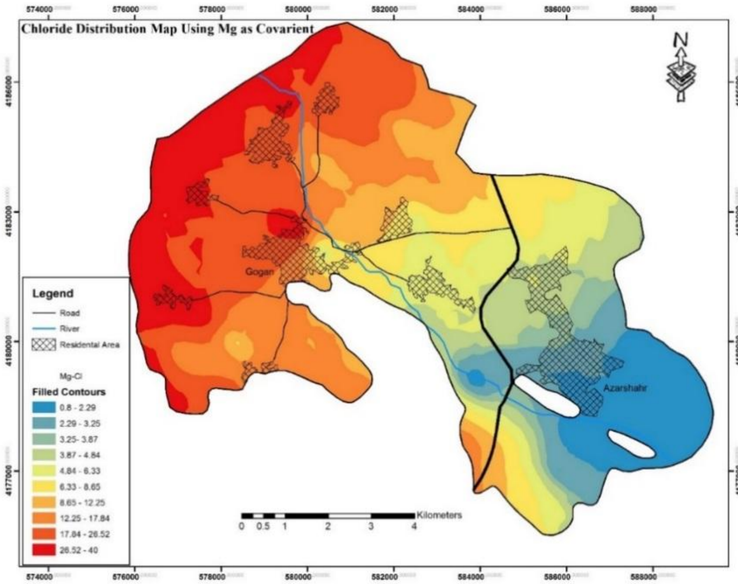


Fig .12. Prediction Map of Cl^{-} concentration using Mg^{++} as covariant.

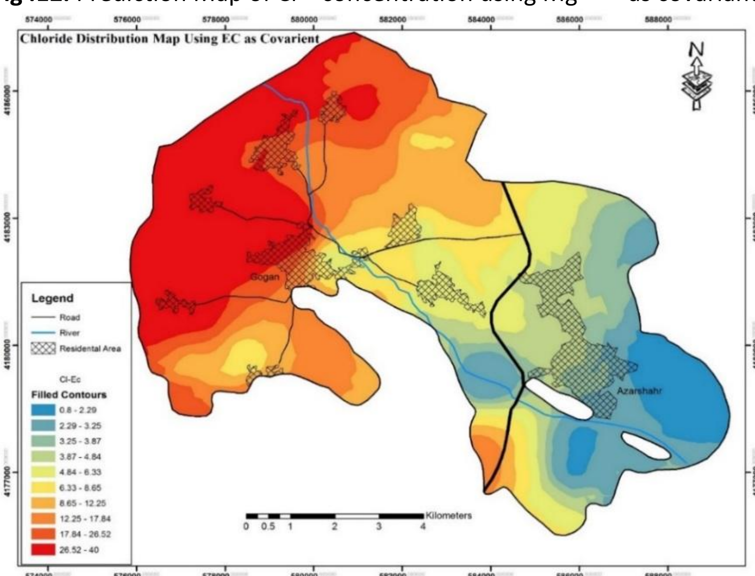


Fig. 13. Prediction Map of Cl^{-} concentration using EC as covariant.

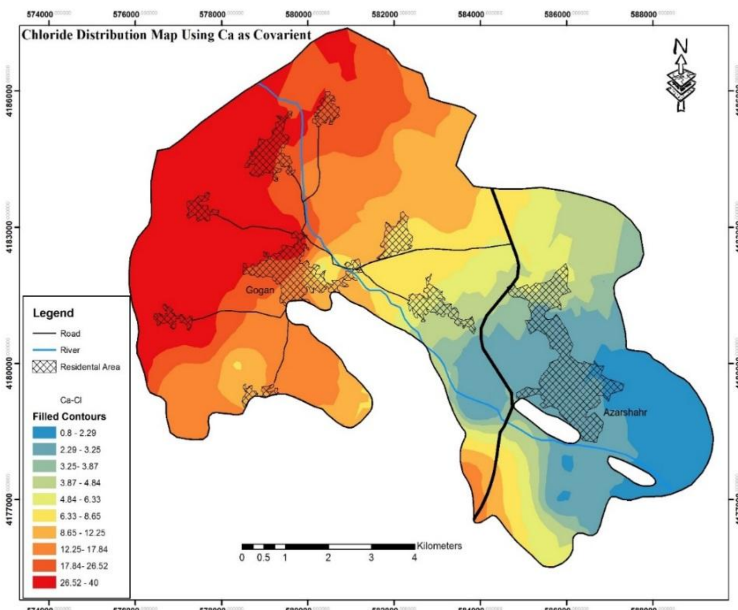


Fig. 14. Prediction Map of Cl^{-} concentration using Ca^{++} as covariant.

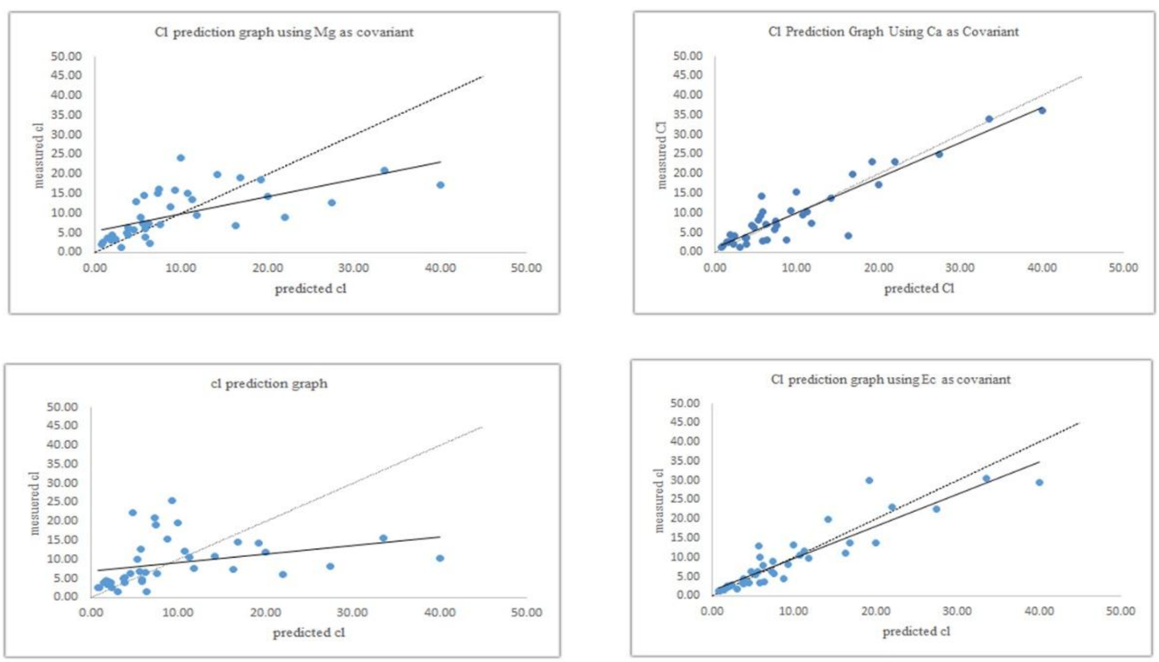


Fig 15. Computed Cross Validation of Cl^{-} concentration using EC, Ca^{++} and Mg^{++}.

Table 4

The Mean Error and RMSE Using EC, Ca^{++} and Mg^{++} as Covariant.

Covariant	Semivariogram/crosscovariance	trend	Lag size	Anisotropy	MEAN ERROR	RMSE
Nothing	Exponential	second	385.70	yes	0.42	2.64
Mg	Exponential	second	259.56	yes	0.22	1.38
EC	Exponential	second	254.60	yes	0.23	1.42
Ca	Exponential	second	412.68	yes	0.04	0.26

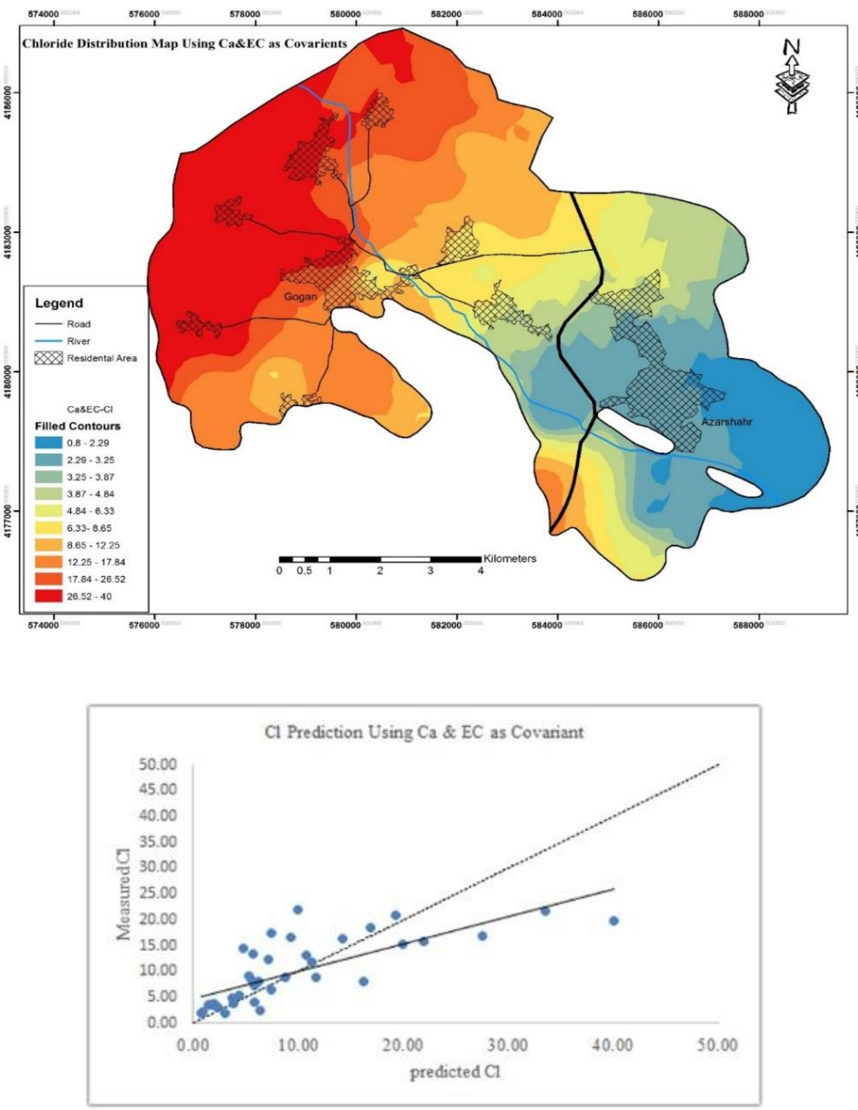


Fig. 16. Computed Cross Validation and Prediction Map of Cl^{-} concentration conjugating EC, Ca^{++} as covariates.

Table 5

The Mean Error and RMSE Conjugating EC and Ca^{++} as Covariant.

Covariant	Crosscovariance	Trend	Lag size	Anisotropy	MEAN ERROR	RMSE
Ca & EC	Exponential	second	259.65	yes	0.32	1.98

4. Conclusion

The results of study revealed that in spatial distribution models based on geostatistic, lognormal ordinary cokriging (LnOCK) usually gives more reliable results than lognormal ordinary cokriging (LnOCK). Also outcomes of research have shown that when covariant has better correlation with unknown parameter, the Error of modelling could be lower. Although adding more data set as excess covariant reduces the model accuracy. Also current study revealed that in areas with less dense sampling, using of spatial prediction via geostatistics can be beneficial for distribution unknown variables estimation, gives an accurate wisdom of the study area. Derived maps by geostatistic finally depicted that, Chloride concentration rises from the South- East to North- West that maybe proves the infiltration of Chloride from the salt pan next to the Aquifer and also Calcium derives from the Travertine member and replaces with Sodium.

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