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# Predicting the spatial distribution of sulfate concentration in groundwater of Jampur-Pakistan using geostatistical methods

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#### ABSTRACT

In this study, we investigate the spatial distribution of sulfate concentration in groundwater of tehsil Jampur, Pakistan using geostatistical techniques. Sulfate concentration in drinking water causes chronic diseases like stomach disorder, diarrhea, laxative effects, and food poisoning in human beings, particularly in infants. First, 30 water samples were collected with their spatial coordinates to evaluate the spatial variation and distribution of sulfate concentration in groundwater of tehsil Jampur. Then, we evaluated the assumptions of normality and autocorrelation in the spatial data and used Matern covariance model to assess the correlation structure of response variable (random field). Furthermore, we applied ordinary least square and weighted least square to estimate the variogram parameters. Two interpolation methods, Ordinary Kriging and Bayesian Kriging were used to predict the unmonitored locations within the studied domain. Performance of the interpolation methods was assessed through leave-one-out cross validation. The predictive maps showing results of both the methods are expected to be helpful to the administrative policy-makers in providing safe drinking water.

Keywords: Spatial mapping; Variogram; Health effects; Isotropy; Bayesian Kriging

# 1. Introduction

Pure water is the basic need of human beings and other species to survive in this world. However, contaminated water causes serious health issues. Pakistan is among the countries where majority of the people do not have access to pure drinking water. Pakistan ranks at number 80 among 122 nations regarding drinking water quality [1] where about 65% of the population has access to pure drinking water, while remaining 35% has insufficient water supply and consequently, they are drinking contaminated water [2–6]. As a result, there is an increased morbidity and mortality rate because of drinking unsafe and polluted water [7]. This growing shortage of fresh water is due to improper disposal of waste materials, food residue, industrial waste in water bodies, and improper use of agrochemicals in agriculture [6,7].

Sulfate is a physiochemical parameter that naturally occurs in drinking water [8]. Water moves through the formation of soil and rocks that contain particular sulfate minerals and some of the minerals dissolve in the groundwater. When sulfate concentration in drinking water exceeds 250 mg/l, a medicinal

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taste makes the water unpleasant [8–10]. Mixture of sulfate in drinking water has raised many health issues, especially diarrhea and dehydration in infants and adults [11,12]. Also, people suffer from the laxative effects when they change drinking water from low sulfate concentration to high sulfate concentration. Therefore, many studies have been conducted to see the effects of sulfate on humans, see [10–15]. Water containing sulfate concentration from 250 to 500 mg/l is free from severe health risks [16–18].

Geostatistics offer multiple techniques to measure and predict the pollutant concentration of groundwater and its chemistry in space. A number of researches have been done in past using Geostatistical approaches for modeling and interpolation of water contamination parameters, see [16,17,19-21] and [22,23]. Geostatistics is based on the presence of spatial autocorrelation and requires the normal distribution of response variable. Pilz and Spöck [24] suggested to use the Box-Cox transformed Gaussian response variable when the normality assumption is violated. Hussain et al. [16] remarked that Kriging accounts for the variability of spatial response variable; therefore, better than classical methods. They suggested to evaluate the performance of Kriging through the Kriging variance which depends upon spatial variation of response variable. Talaee [20] used Ordinary Kriging (OK) and Indicator Kriging to examine the spatial variability in groundwater of Ardabil Plain in the northwest of Iran. He analyzed 11 water parameters including sulfate concentration and created predictive maps using the results of OK. Bayesian Kriging (BK), introduced by Omre and Halvorsen [25], is an efficient technique for interpolation of random field at unobserved locations. It takes into account the prior information while estimating the variogram parameters and considers the variogram parameters as a random variable [24-26]. Mubarak et al. [17] studied the spatial distribution of sulfate concentration in drinking water of three divisions of Punjab, Pakistan using OK, BK, and Universal kriging. They finally used spherical variogram model estimated by maximum likelihood estimation method and concluded that BK has least prediction error as compared to other Kriging methods. Roy and Hossain [26] adopted Bayesian geostatistical model to predict the arsenic concentration at different levels of well depth in Bangladesh with the help of spatial predictive maps. They used Matern correlation function and concluded that this function is positive semi-definite and decreases when distances increase. Cressie [27] suggested to use weighted least square (WLS) for the estimation of variogram parameters because WLS takes into account the correlation between variogram estimators at different lags and automatically gives more weight to initial lags and less weight to those having smaller number of pairs. Finley and Banerjee [28] reported that correlation parameters are often weakly identifiable; therefore, selection of proper priors is necessary in BK to run efficient Monte Carlo Markov Chain (MCMC). Roy and Hossain [26] reported that the priors of inverse gamma distribution are suitable for partial sill ( $\sigma^2$ ) and nugget effect ( $\tau^2$ ), while priors of uniform distribution are good choice for range or decay parameter ( $\phi$ ).

The objective of this study is to predict the spatial distribution of sulfate concentration in groundwater of tehsil Jampur, Pakistan. For this purpose, we examined the assumption of normality and spatial autocorrelation of response variable [24,29]. The null hypothesis of no spatial dependence between observed locations was tested through nugget to sill ratio [30]. We estimated the parameters of variogram model by OLS and WLS [27]. Two interpolation methods, OK and BK, have been applied with Matern variogram model to predict the ungauged locations and prediction results have been displayed using contour plots. Finally, both OK and BK methods have been cross validated using leave one out cross validation [31] and technique with better performance is suggested for further prediction.

#### 2. Material and methods

## 2.1. Study area and data description

The study area, tehsil Jampur is located in southern Punjab, Pakistan, at east latitude 29.38° and north longitude 70.35°. Jampur is tehsil headquarter situated in the north of district Rajanpur and about 45 km away from Dera Ghazi Khan. It lies 15 km away from the west bank of Indus River and occupies an area of approximately 10 km<sup>2</sup> with population about 120,000. Jampur has a desert climate with hot summer while mild winter [23]. Furthermore, average maximum temperature of Jampur varies between 26.0 and 44.0°C. Except July-August (average approximate precipitation is 55 mm), the weather is generally dry throughout the year. It has much spatial variability in groundwater and it was found that majority of its population was drinking contaminated water [32]. Therefore, a survey was conducted to evaluate the quality of groundwater. Keeping in view the guidelines of World Health Organization (WHO), 30 samples of groundwater were collected from different locations (see Fig. 1). For a thorough and objective analysis, 9 samples were taken from injector pump, 15 from hand pump, 2 from water supply, and 4 from tube well. For



Fig. 1. Location map showing the spatial distribution of monitored places of tehsil Jampur, Pakistan. Source: [32].

each sample, groundwater was processed and analyzed for the sulfate concentration (mg/l). Spatial coordinates of each location were observed using handheld GPS receiver and were recorded in GCS system. Sulfate concentration (mg/l) was observed using sulfate Portable Photometer Hi 96751 HANNA Instrument USA. The methodology of Gilcreas [33] was implemented to observe the sulfate level in sampled locations for modeling and prediction purpose.

#### 2.2. Geostatistical analysis

# 2.2.1. Variogram model

Variogram is a tool used to quantify the spatial variability and correlation structure in response

variable. It reflects our understanding about the geometry and continuity of response variable and determines that how data values are related with distances [27,34]. Mathematically, semi variogram is described as:

$$\gamma(h) = \frac{1}{2|N(h)|} \sum_{i=1}^{N(h)} [Y_i - Y_j]$$
(1)

where |N(h)| shows the total number of Euclidean distances separated by lag (*h*) and  $Y_i$ ,  $Y_j$  are data values at spatial locations *i*, *j*, respectively. Variogram is mainly used to determine the spatial variation in random field (response variable). Therefore, selection of suitable variogram model plays significant role in spatial prediction [21]. Roy and Hossain [26] suggested to use the Matern variogram model because it decreases when distances increase and according to Finley and Banerjee [28], it takes the mathematical form:

$$\gamma(h) = \begin{cases} \tau^2 + \sigma^2 [1 - (1 + \phi h)e^{-\phi h}], & h > 0\\ 0, & h \le 0 \end{cases}$$
(2)

where  $\tau^2$  is the nugget effect,  $\sigma^2$  is sill, and  $\phi$  is the range of the Matern model. The lag distance is defined as the distance between pairs at which the variogram is calculated, while the distance beyond which spatial autocorrelation is practically zero is the range ( $\phi$ ). The nugget effect ( $\tau^2$ ) represents the micro scale variation or measurement error [26].

# 2.3. Theory of Kriging

Efficient interpolation method helps in reducing prediction error [16,17,21]. Kriging is the best linear unbiased prediction method [35]. The predicted value at unmonitored location using Kriging method is a linear combination of observed locations. Therefore, to determine the coefficient of the linear combination, auto covariance plays a significant role [29]. In OK, mean is assumed constant in the local neighborhood of each estimation point i.e.  $m(\mu_{\alpha}) = m(\mu)$ , for each adjacent data value,  $(\mu_{\alpha})$  is used in estimating  $Y(\mu)$ . Kriging estimator takes the form:

$$Y^{*}(\mu) = m(\mu) + \sum_{\alpha=1}^{n(\mu)} \lambda_{\alpha}(\mu) [Y(\mu_{\alpha}) - m(\mu)]$$
(3)

$$Y^*(\mu) = \sum_{\alpha=1}^{n(\mu)} \lambda_\alpha(\mu) \ Y(\mu_\alpha) + \left[1 - \sum_{\alpha=1}^{n(\mu)} \lambda_\alpha(\mu)\right] m(\mu) \tag{4}$$

where  $\lambda_{\alpha}$  is representing the weights associated with Kriging estimator given in Eq. (3). Further, unknown mean is shifted in such a way that kriging weights sum to unity. Consequently, estimator of OK becomes:

$$Y_{\text{OK}}^*(\mu) = \sum_{\alpha=1}^{n(\mu)} \lambda_{\alpha}^{\text{OK}}(\mu) Y(\mu_{\alpha}) \quad \text{with} \quad \sum_{\alpha=1}^{n(\mu)} \lambda_{\alpha}^{\text{OK}}(\mu) = 1 \quad (5)$$

According to the constraint of unit sum of weights, error variance is minimized by arranging the system that reduces the error variance plus an additional term involving a Lagrange's parameter,  $\mu_{OK}(\mu)$ :

$$L = \sigma_{\rm e}^2(\mu) + 2\mu_{\rm OK}(\mu) \left[ 1 - \sum_{\alpha=1}^{n(\mu)} \lambda_\alpha(\mu) \right]$$
(6)

The reduction in error variance with respect to Lagrange's parameter  $\mu_{OK}(\mu)$  effects the constraint in such a way that it becomes:

$$\frac{1}{2}\frac{\partial L}{\partial \mu} = 1 - \sum_{\alpha=1}^{n(\mu)} \lambda_{\alpha}(\mu) = 0 \tag{7}$$

Thus, from Eq. (7), it is clear that unbiasedness property of Kriging estimator hold as:

$$\sum_{lpha=1}^{n(\mu)} \lambda_{lpha}(\mu) = 1$$

Afterward, the error variance  $(\sigma_{OK}^2(\mu))$  is deduced from Kriging equations as given below in Eq. (8):

$$\sigma_{\rm OK}^2(\mu) = c(0) - \sum_{\alpha=1}^{n(\mu)} \lambda_{\alpha}^{\rm OK}(\mu) \ C(\mu_{\alpha} - \mu) - \mu_{\rm OK}(\mu)$$
(8)

Here the covariance function c(0) utilizes the vectorvalued of parameter  $\alpha$  where  $\alpha = c(\sigma^2 = \text{sill}, \phi = \text{range}, \tau^2 = \text{nugget})$ .

BK introduced by Omre and Halvorsen [25] is considered as the best prediction technique [34] because it takes into account the parameters uncertainty for estimating the variogram parameters. To normalize the non-normal response variable, Box–Cox transformation [36] is used as:

$$Y^{\text{transformed}} = \begin{cases} \frac{Y^{\lambda} - 1}{\lambda} : & \lambda \neq 0\\ \log(Y) : & \lambda = 0 \end{cases}$$
(9)

where  $\lambda$  is the transformation parameter known as lambda. Box–Cox transformation is implemented on a response variable having only positive values [36]. For detailed analysis of both OK and BK, we used *geoR* package [31] of R statistical software [37]. To carry out spatial prediction using OK, the *krige.control* function is implemented which is further used in *krige.conv* function. Initially, as input parameters, the data and spatial coordinates were defined by *geodata* argument and specified the locations where prediction was required.

#### 2.4. Assessing the presence of anisotropy

Spatial autocorrelation in terms of spatial anisotropy or isotropy is a basic feature in spatial modeling for natural phenomenon [35]. A correlation structure is isotropic when the pattern of the spatial correlation changes due to change in the direction of orientation of pairs of locations. Kriging methods are based on isotropic models [34]. Therefore, the correction for any anisotropy is necessary before using Kriging approaches. Usually the presence of spatial anisotropy is assessed by directional variogram maps and anisotropy ratio [29]. We found using function *coords.aniso* that the structure of spatial autocorrelation of response variable was approximately independent from direction. Therefore, omnidirectional variogram (Fig. 5) was used to capture autocorrelation.

#### 2.5. Criteria for performance evaluation

To compare the quality of prediction obtained from fitted OK and BK techniques, we used four performance evaluation measures: root mean square error (RMSE), mean absolute error (MAE), relative bias (rBIAS), and relative mean squared prediction error (rMSEP) [38]. Mathematically these are expressed as:

RMSE = 
$$\sqrt{\frac{1}{k} \sum_{i=1}^{k} (\hat{Y}_i - Y_i)^2}$$
 (10)

$$MAE = \frac{1}{k} \sum_{i=1}^{k} \left| \hat{Y}_i - Y_i \right|$$
(11)

$$\text{rBIAS} = \frac{1}{k\overline{Y}} \sum_{i=1}^{k} (\hat{Y}_i - Y_i)^2 \tag{12}$$

$$\text{rMSEP} = \sum_{i=1}^{k} (\hat{Y}_i - Y_i)^2 / \sum_{i=1}^{k} (\overline{Y}_p - Y_i)^2$$
(13)

 $\hat{Y}_i$  shows the predicted values at index *i* and  $\overline{Y}$  is quantity of arithmetic mean while  $\overline{Y}_p$  is the arithmetic mean of predicted values. We used leave-one-out cross validation where one data location is removed from data-set and it is predicted from the remaining data [31].

#### 3. Results and discussion

#### 3.1. Exploratory data analysis

The measured sulfate concentration at sampled locations was analyzed by R statistical software [37]. Since, most of the geostatistical techniques depend upon the normality assumption of response variable [29]; therefore, at first step, the normality and spatial autocorrelation assumptions of spatial data are evaluated. Sulfate concentration in our data-set ranged from 45 to 3,700 mg/l. The level of observed sulfate concentration in groundwater is shown in Fig. 2.

The mean, median, mode, and standard deviation (SD) of sulfate concentration were 731.4, 401, 3,700, and 855.68 mg/l, respectively, which showed that the distribution of sulfate concentration is positively skewed. To test the normality of response variable,  $(A^2 = 2.22,$ Anderson–Darling Normality test, *p*-value < 0.005), confirmed the departure from normality. Further, the histogram in Fig. 3 also indicated that response variable is far from normality and is positively skewed with approximately 80% of the samples in the lower tail while only six samples exceed the limit of 1,000 mg/l. To normalize the skewed data, Box-Cox transformation [36] implemented by Eq. (9), gave transformed parameter,  $\lambda = 0.065$ , used to transform the skewed response variable. Box-Cox transformed variable takes the form:

Transformed variable = 
$$[{\text{Original variable}}^{0.065}] - 1/(0.065)$$
 (14)

After implementing the Box–Cox transformation, Anderson–Darling normality test was again used to evaluate the normality of transformed variable which took the value of statistic,  $A^2 = 0.393$  with *p*-value = 0.3558, that confirmed normality. Thus, the response variable becomes Box–Cox transformed Gaussian response variable [24].

The next step consists of examining the spatial autocorrelation among the observations of response variable. For this purpose, variogram envelope (Fig. 4)



Fig. 2. Plot of observed sulfate concentration at 30 sampled locations corresponding to spatial coordinates.



Fig. 3. Exploratory data analysis (EDA) of observed sulfate concentration (mg/l) of Jampur Region, showing positively skewed distribution.

that contains an envelope which is based on the permutations calculated from data values across the sampled locations. Variogram envelope is designed under the null hypothesis:  $H_0$  = No spatial autocorrelation. It showed that the increasing trend in the experimental variogram is statistically significant because all points are lying within limits; therefore, data follow the basic assumption of spatial autocorrelation [29]. Further, Geostatistics is also based on an assumption that the attributes in the earth are spatially continuous up to certain lag distance [29,34] which is evaluated by perspective plot [31] shown in Fig. 4 (right panel).

# 3.2. Parameter estimation of experimental variogram

An important step in geostatistical prediction is the selection of suitable correlation structure to model the variogram and its parameters estimation [16]. Therefore, three correlation structures like exponential, spherical, and Matern are fitted over the experimental variogram model (Fig. 5). Their estimated parameters using *eyefit* command of geoR package [31] are  $(\sigma^2 = 3.12, \phi = 0.26, \tau^2 = 2.21)$  for exponential,  $(\sigma^2 = 3.34, \phi = 0.24, \tau^2 = 2.08)$  for spherical, and  $(\sigma^2 = 3.12, \phi = 0.27, \tau^2 = 2.08)$  for Matern variogram model with smoothness parameter,  $\gamma = 0.27$ . Among three variogram models, Matern variogram model gave least value of RMSE. Therefore, the estimated

parameters of Matern variogram model are used in Kriging. The lag value for experimental variogram was 46.8 meters calculated by Eq. (1) (Matheron Estimator) [30]. Parameters of selected Matern variogram model have been estimated by OLS and WLS. In final prediction, the parameters estimated by WLS were used as it produced less RMSE (Table 1). The parameters estimated using WLS showed that the ratio of  $\tau^2/\sigma^2$  was approximately 62%, representing that the spatial autocorrelation of sulfate concentration was moderately dependent. It also showed that the sulfate concentration differ at a scale smaller than the lowest lag distance [39].

# 3.3. Kriging estimation and prediction maps

The estimated values of covariance parameters, partial sill ( $\sigma^2$ ), and range ( $\phi$ ), were defined as a 2 element vector (3.12, 0.27) in *cov.par* argument. Moreover, the type of adopted correlation function, Matern, was mentioned in *cov.model* argument. The results of prediction mean and prediction SD were shown (see Figs. 5 and 6 (left panel)) using *image* function. In BK, model specifications were same as described in OK [17]. Moreover, for BK, prior distributions were considered for each uncertain parameter of the models because Finley and Banerjee [28] reported that for an efficient MCMC behavior, reasonable priors for



Fig. 4. Plot of variogram envelope of sulfate concentration to examine spatial autocorrelation between neighboring samples (left panel). Perspective plot showing the continuity assumption of response variable over study domain (right panel).



Fig. 5. Effect of exponential, spherical and Matern variogram models (with kappa = 0.27) over the experimental variogram model fitted by WLS estimation method.

{ $\tau^2$ ,  $\phi$ ,  $\sigma^2$ , v} are required. For this purpose, sample variogram (Fig. 5) is a useful tool [26]. For range ( $\phi$ ), the priors of uniform distribution were taken with hyper parameters, 1.0 and 0.5. Commonly, the priors of inverse gamma distribution are assigned to variance

components, partial sill ( $\sigma^2$ ) and nugget ( $\tau^2$ ) [26]. Therefore, the prior distribution for nugget effect  $(\tau^2)$ , was taken as inverse gamma with scale and shape parameter 2 and 1.5, respectively. Similarly, for partial sill ( $\sigma^2$ ), inverse gamma distribution with scale and shape parameter 2 and 1.5 was assumed as a prior distribution. Uniform distribution was specified as a prior distribution for nugget to sill ratio ( $v = \tau^2/\sigma^2$ ). To obtain the posterior distribution of parameters, 5,000 iterations of MCMC were executed and the convergence of the fitted models was observed. The function krige.bayes of geoR package [28] was used while implementing BK. Like OK, the results obtained from BK were also displayed for prediction mean and prediction SD (Figs. 5 and 6 (right panel)). Comparatively, BK showed a good picture of sulfate concentration in groundwater and gave lower prediction error (see Table 2).

Estimated values of OK and BK are plotted using image plots as shown in Fig. 6 (Mean Predictive Maps) and Fig. 7 (Prediction SD Maps). Kriging estimates showed different spatial patterns and identified high risk areas. The higher values have been indicated

Table 1 Parameters estimation of Matern variogram model using OLS and WLS

Estimation technique	Range (ø)	Partial sill ( $\sigma^2$ )	Nugget ( $\tau^2$ )	RMSE
OLS	0.241	3.1412	2.092	5.683
WLS	0.255	3.3514	2.085	4.129



Fig. 6. Image plots of interpolated sites using BK (right panel) and OK (left panel).

Table 2							
Results	of	various	spatial	validation	statistics	for	Ordinary
and BK							

	Various spatial validation statistics				
Kriging technique	RMSE	MAE	rBIAS	rMSEP	
OK BK	949.94 865.99	620.33 614.76	0.3449 0.1550	7.1292 3.4531	

by blue, yellow, and red colors which are showing hotspot areas while tolerable values have been showed by green color, representing good region. Further, the highest sulfate concentration (greater than 2,000 mg/l) is observed at top left area of prediction map with east latitude 29.60–29.63° and north longitude 70.33–70.39°. The permissible limits as described by WHO [18] are given at region with east latitude 29.52–29.60° and north longitude 70.30–70.45° as shown in Fig. 6 (right panel). Similarly, spatial region with east latitude



Fig. 7. Image plots of prediction SDs using BK (right panel) and OK (left panel).

29.45–29.70° and north longitude 70.55–70.60° showed permissible level. BK showed relatively lower prediction SD as compared to OK which is clearly shown in Fig. 7.

# 3.4. Cross validation for assessing performance of predicted errors

Leave-one-out cross validation is used where each of the data location is removed one by one and then predicted using the remaining data [31]. Both, OK and BK have been compared using the validation approaches and the resulted predicted errors have been given in Table 2.

These statistics are calculated using package *spTimer* [38] of R statistical software [37]. The results taken by cross validation (Table 2) illustrate that BK gives much better estimation results than OK, because all validation statistics: RMSE, MAE, rBIAS, and rMSEP predicted by BK are smaller than RMSE, MAE, rBIAS and rMSEP predicted by OK. Thus, results showed the better performance of BK for the prediction of groundwater salinization at unmonitored locations.

# 4. Conclusion

This study aimed to elaborate the use of geostatistical interpolation methods for mapping the level of sulfate concentration in ground drinking water. BK gives better prediction and valid estimation of sulfate concentration in groundwater as compared to OK. Our findings confirm the results reported in [17]. Moreover, the highest sulfate concentration (greater than 2,000 mg/l) estimate is observed at east latitude 29.60–29.63° and north longitude 70.33–70.39°. As suggested by Cressie [27], our results also support the use of WLSs method for the estimation of variogram parameter.

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