**Abstract**

Exposure of high concentration of ground-level ozone (GLO) can trigger a variety of health problems including chest pain, coughing, throat irritation, asthma, bronchitis and congestion. There are substantial human and animal toxicological data that support health effects associated with exposure to ozone and associations have been observed with a wide range of outcomes in epidemiological studies. The aim of the present study is to estimate the spatial distributions of GLO using geostatistical method (ordinary kriging) for assessing the exposure level of ozone in the eastern part of Texas, U.S.A. GLO data were obtained from 63 U.S. EPA’s monitoring stations distributed in the region of study during the period January, 2012 to December, 2012. The descriptive statistics indicate that the spatial monthly mean of daily maximum 8 hour ozone concentrations ranged from 30.33 ppb (in January) to 48.05 (in June). The monthly mean of daily maximum 8 hour ozone concentrations was relatively low during the winter months (December, January, and February) and the higher values observed during the summer months (April, May, and June). The higher level of spatial variations observed in the months of July (Standard Deviation: 10.33) and August (Standard Deviation: 10.02). This indicates the existence of regional variations in climatic conditions in the study area. The range of the semivariogram models varied from 0.372 (in November) to 15.59 (in April). The value of the range represents the spatial patterns of ozone concentrations. Kriging maps revealed that the spatial patterns of ozone concentration were not uniform in each month. This may be due to uneven fluctuation in the local climatic conditions from one region to another. Thus, the formation and dispersion processes of ozone also change unevenly from one region to another. The ozone maps clearly indicate that the concentration values found maximum in the north-east region of the study area in most of the months. Part of the coastal area also showed maximum concentrations during the months of October, November, December, and January.

**Key words:** Ground level ozone (GLO), Geostatistics, Kriging, Mapping

**1. Introduction**

Tropospheric Ozone (O₃) is listed by many countries and organisation like the World Health Organization (WHO) and United States Environmental Protection Agency (U.S. EPA) as one of its criteria pollutants. It is a major constituent of photochemical smog, an air pollution event that often occurs in urban areas. Ozone is proven to have adverse effects on human health (Lippmann, 2009; Bascom et al., 1996). It can also adversely affect crops and forest ecosystems (Bassin et al., 2007; Hayes et al., 2007; Mills et al., 2007; Schaub et al., 2005; U.S. EPA, 1996; Fuhrer, 1994; Sanders et al., 1994). Due to its association with health, the study of ground level ozone has become the target of several researchers in recent years. With a view to deduce more accurate predictive models, different approaches were used for the mapping of ground level ozone. However, literature survey shows that studies about the temporal and spatial variability of ground level ozone are very limited.

The mapping of ground level ozone in urban areas assists the policy makers to describe and quantify the pollution at locations where no measurements has been done. The preparation of ozone maps is feasible, if a spatial correlation of the variable of interest is identified (Hopkins, 1999). The existence of a spatial correlation of ozone is not only a condition for an optimum interpolation of the data in space in order to generate a map of ozone, but it also provides very useful

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**Kriging Analysis for Spatio-temporal Variations of Ground Level Ozone Concentration**

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insights on the formation and distribution processes.

Many air pollution studies have employed distance-weighting methods (e.g., Phillips, 1997), but kriging is the only one that incorporates the spatial correlation into its estimation algorithm. Kriging has been used more widely (Gorai et al., 2013; Maity, 2006; Sarangi et al., 2006; Araghi nejad and Burn, 2005; McGrath, 2004; Juang et al., 2002; Germann and Joss, 2001; Gringarten and Deutsch, 2001; Lin et al., 2001; Merino et al., 2001; Tayanc, 2000; Poon et al., 2000; Tran chant and Vincent, 2000; Yamamoto, 2000; Kravchenko and Bullock, 1999; Gotway et al., 1996; Hosseini and Gallichand, 1994) in environmental variable mapping, due to its many advantages (Goovaerts, 1997). Although kriging requires an abundance of sample points to be an accurate spatial interpolation method (Myers, 1991), even when relatively small data sets and not exhaustive samplings are available it is a reliable technique for investigating the distribution and sources of pollutants (Carlon, 2001).

The spatial distributions of GLO concentration have some heterogeneity and the concentrations are rarely available for every possible location in an area. The measurement of GLO concentration at every location is not always feasible in view of the time and the cost involved in data collection. Therefore, prediction of values at other locations based upon selectively measured values could be one of the alternatives. Therefore, data samples are transformed via a series of interpretation steps to acquire complete descriptions of phenomena of interest (Edwards et al., 2001). A geostatistical scheme is a regular procedure that is an efficient way of mapping according to the stochastic spatial variation. The basic assumption in using geostatistics is that the properties in the atmosphere have some spatial continuity (Vardoulakis, 2005; Coppalle, 2001) up to a certain lag distance. The geostatistical concepts and its applications are reported by different researchers around the world (Webster and Oliver, 2007; Kumar and Ahmed, 2003; Goovaerts, 1997; Isaaks and Srivastava, 1989; Journel and Huijbregts, 1978). Kriging is a geostatistical method consisting of a linear interpolation approach that provides a best linear unbiased estimator for quantities that vary spatially (Goovaerts et al., 1997; Keefer, 1994; Isaaks et al., 1989). Moreover, besides interpolation, kriging provides information on interpolation errors. Such values can be mapped to generate error surfaces which inform about the reliability of estimates. Kriging is divided into two distinct tasks: viz. quantifying the spatial structure of the data and producing a predicted surface. In order to predict an unknown value for a specific location, Kriging will use the fitted model from variography, the spatial data configuration, and the values of the measured sample points around the prediction location (Sarangi et al., 2006). It makes good use of existing knowledge by considering the difference of an attribute varies in space through the variogram model (Webster, 2007). It interpolates algorithms to generate maps of the best local estimate and generally smooths out the local details of the spatial variation of a particular attribute (Lin et al., 2009). Kriging method considers the spatial correlation between the sample points, and is mostly used for spatial variability mapping (Ella et al., 2001; Stein, 1999). Kriging is distinguished from an inverse distance weighted (IDW) and other interpolation methods by taking into consideration the variance of estimated parameters (Buttner et al., 1998).

The objective of the present study is to understand spatiotemporal changes of GLO concentrations in the eastern region of Texas State, United States. The present study also examined the different kriging models for minimizing the prediction error.

## 2. MATERIALS AND METHODS

### 2.1 Study Area

Eastern part of Texas State, U.S.A (shown in the Fig. 1) was considered for the distribution analysis of ozone concentration. Due to an insufficient number of monitoring stations in the western part of the state, only eastern part was considered for the study. Texas State located in the west-south-central region of the United States. The longitude and latitude of the state are ranged from 93°31′W to 106°38′W and 25°50′N to 36°30′N respectively. It is the second most populous (25,145,561), and 29th most densely populated (96.3 inhabitants per square mile of land area) state of the 50 United States (U.S. Census Bureau: Resident Population Data, 2010). Texas covers 696, 241 square kilometers of land area and ranks as the 2nd state by size (U.S. Census Bureau, State Area Measurement). Texas is bordered on the north by Oklahoma and Arkansas (with part of the line formed by the Red River); on the east by Arkansas and Louisiana (with part of the Louisiana line defined by the Sabine River); on the south-east by the Gulf of Mexico; on the south-west by the Mexican states of Tamaulipas, Nuevo León, Coahuila, and Chihuahua (with the line formed by the Rio Grande); and on the west by New Mexico. In general, the climate in Texas State varies widely, from arid in the west to humid in the east. There is significant variation in the geography from one region to another in Texas State. There are coastal regions, mountains, deserts and wide-open plains. In coastal regions, the weather is neither particularly hot in the summer nor particularly cold during the winter. East Texas has the humid subtropical cli-
mate typical of the Southeast, occasionally interrupted by intrusions of cold air from the north.

2.2 Ozone Concentration Data

The ozone concentration data collected by U.S. EPA’s Air Quality System (AQS) at the various monitoring stations located in different counties of eastern part of Texas for the year 2010 were used for the study. The data used in this study were taken from the United States Environmental Protection Agency (U.S. EPA) air quality system data mart (Source: http://www.epa.gov/airdata/ad_rep_mon.html). The data were obtained for 63 monitoring stations distributed in the study area. The characteristics of the raw data collected from the website are daily maximum 8 hours average concentrations of ozone. The daily data for each monitoring station were used for determination of monthly average concentrations.

2.3 Geostatistical Method (Kriging)

Geostatistics is a class of statistical techniques developed to analyze and predict values of a species distributed in space or earth. It begins with a type of autocorrelation analysis called semivariance analysis, in which the degree of spatial self-similarity is displayed as a variogram. Several different forms of kriging (simple, ordinary, indicator, universal, disjunctive and probability) were developed. With simple kriging, one assumes that the mean value is known and constant, while with ordinary kriging the mean value is determined during the interpolation. Indicator kriging assumes that the mean is constant and unknown. For non-stationary variation, where the data follow a trend, universal kriging or kriging with intrinsic random functions are used (Mulholland et al., 1998; Oliver, 1996). We adopted ordinary kriging for all the interpolations for O3. In the present study, ordinary kriging estimations were performed based on the fitted spherical semivariogram models. Spherical semivariogram models were fitted using 63 samples distributed in the area in each of the 12 months. Geostatistical analysis was conducted with the following steps:

2.3.1 Exploratory Data Analysis

Exploratory data analysis was performed to check data consistency, removing outliers, and identifying...
Fig. 2. Q-Q Plots for average monthly average data (a) January (b) February (c) March (d) April (e) May (f) June (g) July (h) August (i) September (j) October (k) November (l) December.
statistical distribution. It is important to detect outliers because they may be the values that were measured or recorded incorrectly and, in this case, their effects on subsequent stages of the geostatistical study are very negative. Kriging methods work best for normally distributed data (Goovaerts et al., 1997). Transformations can be used to make the data normally distributed and satisfy the assumption of equal variability in the data. The normal Q-Q plots were derived using SPSS software version 21. The plots of each of the twelve months are represented in Fig. 2(a)-(l) to check the normality of the observed data on monthly average ozone concentrations. Q-Q Plots clearly indicate that the data for each month are closely followed a normal distribution. The normalcy of the data was further checked by statistical analyses (descriptive statistics).

Table 1 summarizes the descriptive statistics of average monthly ozone concentrations for the year 2012. The statistical analysis of ground level ozone concentration data of Texas, U.S.A in the year 2012 indicating that the spatial monthly mean of daily maximum 8-hour ozone concentrations ranges from 30.33 ppb (in January) to 48.05 (in June). The monthly mean of daily maximum 8-hour ozone concentrations was relatively low during the winter months (December, January, and February) and, this may be due to low solar radiation levels received during the season. Similarly, the higher values of monthly mean of daily maximum 8-hour ozone concentrations were observed during the summer months (April, May, and June) due to higher levels of solar radiation. The spatial variation in ozone concentrations was observed high in the months of July (Standard Deviation: 10.33), and August (Standard Deviation: 10.02) indicates the regional variations in geographic and climatic conditions in the area during these months.

The statistical results also showed that the monthly average ozone concentrations varied significantly during the period. Ozone levels are more regularly distributed during the winter months than the summer months. For each sampling campaign, the mean and median are very similar, which is, indicative of data coming from a normal distribution is somewhat nearer. Thus, in the present study, no transformation of data was done for geostatistical analyses.

### 2.3.2 Structural Analysis of Data
Spatial correlation or dependence can be quantified with semivariograms (or variograms). Kriging relates the semivariogram, half the expected squared difference between paired data values \( z(x) \) and \( z(x + h) \) to the distance lag \( h \), by which locations are separated. The basic function of semivariogram model for continuous sampling site is given by equation (1).

\[
\gamma(h) = \frac{1}{2} \mathbb{E}[(z(x) - z(x + h))^2]
\]  

For discrete sampling sites, the function is written in the form of equation (2).

\[
\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [z(x_i) - z(x_i + h)]^2
\]

Where, \( z(x_i) \) is the value of the variable \( z \) at the location of \( x_i \), \( h \) is the lag, and \( N(h) \) is the number of pairs of sample points separated by \( h \). For irregular sampling, it is rare for the distance between the sample pairs to be exactly equal to \( h \). A semivariogram plot is obtained by calculating the values of the semivariogram at different lags. These values are then usually fitted with a theoretical model: circular, spherical, exponential, stable or Gaussian. The models provide information about the spatial structure as well as the input parameters for the kriging interpolation.

Prediction capabilities were tested for different variogram models and found that the results were not changed significantly with a change in the type of variogram model. Thus, the spherical variogram model was used for spatial ozone concentration prediction for each case. The variogram models for twelve months are represented in Fig. 3(a)-(l) and its corresponding characteristic parameters (Sill, Nugget, Range, and Lag value) are listed in Table 2.

### 2.3.3 Predictions and Cross Validation
Spherical semivariogram models were used for prediction of ozone concentration. Predictive performances of the fitted models were checked by cross validation tests. For the cross-validation test, the values of mean error (ME), mean square error (MSE), root mean square error (RMSE), average standard error (ASR) and root

| Table 1. Descriptive statistics of monthly spatial distributions of ozone concentration |
|---------------------------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
|                                 | Jan | Feb | March | April | May | June | July | Aug | Sept | Oct | Nov | Dec |
| Minimum                        | 23.94 | 25.00 | 31.03 | 40.00 | 36.90 | 29.03 | 19.07 | 20.50 | 33.23 | 31.94 | 34.04 | 24.40 |
| Maximum                        | 35.16 | 37.62 | 49.88 | 54.47 | 55.74 | 59.21 | 54.35 | 59.84 | 57.13 | 47.93 | 48.23 | 37.37 |
| Mean                           | 30.33 | 31.01 | 39.74 | 47.93 | 48.03 | 48.05 | 35.19 | 43.17 | 44.41 | 40.76 | 40.25 | 31.37 |
| Median                         | 30.55 | 31.21 | 39.80 | 47.90 | 48.23 | 48.30 | 31.16 | 40.70 | 44.37 | 40.65 | 40.48 | 31.26 |
| St. Dev.                       | 2.70  | 2.80  | 2.92  | 2.93  | 4.72  | 6.28  | 10.33 | 10.02 | 5.21  | 2.73  | 2.70  | 2.81  |
Fig. 3. Variogram models for twelve months (a) January (b) February (c) March (d) April (e) May (f) June (g) July (h) August (i) September (j) October (k) November (l) December.
mean square standardized error (RMSSE) estimated to ascertain the performance of the developed models. If the predictions are unbiased, the ME should be near zero. However, this statistic has some important drawbacks: it depends on the scale of the data and is insensitive to inaccuracies in the variogram. So, usually the MSE is used to standardize the ME, being ideally zero, i.e., an accurate model would have an MSE close to zero. Besides making predictions, each of the kriging techniques gives the kriging variances that estimate the variability of the predictions from the known values. The kriging variances must be accurately calculated because they have an important influence on some applications of kriging, e.g., the probability kriging. If the RMSE is close to the ASE, the prediction errors are correctly assessed. If the RMSE is smaller than the ASE, then the variability of the predictions is underestimated; conversely, if the RMSE is greater than the ASE, then the variability of the predictions is underestimated. The same could be deduced from the RMSSE statistic. It should be close to one. If the RMSSE is greater than one, the variability of the predictions is underestimated; likewise if it is less than one, the variability is overestimated. After conducting the cross validation process, maps of kriged estimates were generated which provided a visual representation of the
distribution of the ozone concentrations. Various errors are defined by the equations (3)-(7) given below. The corresponding error values of the fitted theoretical models were determined and reported in Table 2.

\[
\begin{align*}
\text{ME} &= \frac{1}{N} \sum_{i=1}^{n} [\hat{Z}(X_i) - Z(X_i)] \\
\text{MSE} &= \frac{1}{N} \sum_{i=1}^{n} [\hat{Z}(X_i) - Z(X_i)]/\hat{o}(X_i) \\
\text{RMSE} &= \sqrt{\frac{1}{n} \sum_{i=1}^{n} [\hat{Z}(X_i) - Z(X_i)]^2} \\
\text{ASE} &= \sqrt{\frac{1}{n} \sum_{i=1}^{n} \hat{o}^2(X_i)} \\
\text{RMSSE} &= \frac{1}{n} \sum_{i=1}^{n} [\hat{Z}(X_i) - Z(X_i)]/\hat{o}(X_i))^2
\end{align*}
\]

where \( \hat{o}^2(X_i) \) is the kriging variance for location \( x_i \), \( \hat{Z}(X_i) \) is predicted value and \( Z(X_i) \) is the actual (measured) value at location \( X_i \) (ESRI, 2003; Goovaerts, 1997). All these analyses were carried out using Geostatistical Analyst module of ArcGIS software version 10.2.
Fig. 4. Monthly average spatial distribution of ground level ozone concentrations (a) January (b) February (c) March (d) April (e) May (f) June (g) July (h) August (i) September (j) October (k) November (l) December.
3. RESULTS AND DISCUSSION

Q-Q plot normal distribution test of monthly average ozone concentrations showed that the data were normally distributed except in a few cases. The outlier data were removed before the variogram analysis. To depict patterns of ozone concentration distribution in the study region, experimental variograms and their variogram models were first analyzed for each month in 2012. Table 2 represents the characteristic parameters of spherical fitted semivariogram models of ground level ozone concentrations. The range and the sill are the two most important parameters of the semivariogram model to describe data. For a detailed discussion about the semivariogram models, refer the book “Geostatistics for environmental scientists” by Webster and Oliver (2007). The ratio of the nugget variance to sill expressed in percentages can be regarded as a criterion for classifying the spatial dependence of ground level ozone concentrations. If this ratio is less than 25%, then the variable has a strong spatial dependence; if the ratio is between 25 and 75%, the variable has moderate spatial dependencies and greater than 75%, the variables shows only weak spatial dependence (Shi et al., 2007; Chang, 1998; Chien et al., 1997). In most of the month in 2012, ozone concentrations showed the strong spatial structure except during January to April, October, and December that has a moderate spatial dependence.

The mean standardized errors (MSE) for 12 months (January to December) are found to be –0.011, –0.019, −0.007, −0.008, 0.007, −0.005, −0.010, −0.011, −0.018, −0.034, and −0.013 respectively. The respective values of root mean square standard error (RMSSE) are 0.89, 0.97, 1.02, 0.99, 0.91, 0.97, 0.97, 1.05, 0.92, 1.04, 0.95, and 0.86. The MSE values are close to zero and their corresponding RMSSE values close to one represent a good prediction model. In each case, the corresponding values of RMSE and ASE are close to each other. This also indicates good agreement of the model.

The shape of the variogram was used to understand the spatial structures of ozone concentrations. The sill value was used to quantify the variability of the ozone concentration among the sample sites. The sill (i.e., spatial variation) values in each case were relatively high during the summer months (May to September) in comparison to winter months (January to April and October to December). Also, nugget values (i.e., variability in local areas) do not significantly change with time. There was no specific trend observed in the nugget values. Moreover, the range of the variogram models varies from 0.372 (in the month of November) to 15.59 (in the month of April).

Spatial distribution of monthly average ozone concentrations was conducted through GIS and geostatistics techniques. The monthly average ozone concentration distribution maps produced by the kriging estimations are shown in Fig. 4(a)-4(l). From these maps, it was found that the spatial trends of ozone concentration in the study area were not uniform with the seasons. Thus, the spatial distributions of the area may be dominated by the frequent change in the climatic conditions in different regions. Due to its large geographical boundary and huge variations in the topography the climatic conditions also differ from region to region. Therefore, the formation and dispersion of ozone vary with the regions, and this may be the reason for not showing any uniform spatial trend of ozone concentrations. To identify the exact reasons, region wise climatic conditions and precursor emissions needs to be studied. The distribution map clearly reveals that the low-level concentrations were observed during the months of December, January, and February and these

| Months   | Fitted variogram model | Nugget (C₀) | Partial Sill (C) | Lag size (in degree) | Range (in degree) | [(C₀/(C₀ + C))]| ME   | RMSE | ASE | MSE | RMSSE |
|----------|------------------------|-------------|-----------------|---------------------|------------------|-----------------|------|------|-----|-----|-------|
| January  | Stable                 | 4.077       | 5.680           | 0.1278              | 41.78            | 1.130           | −0.064| 2.311| 2.565| −0.001| 0.89  |
| February | Stable                 | 4.114       | 6.403           | 0.2737              | 39.11            | 2.384           | −0.001| 2.358| 2.385| −0.019| 0.97  |
| March    | Stable                 | 5.245       | 13.40           | 1.037               | 28.31            | 9.336           | −0.094| 2.658| 2.468| −0.011| 1.02  |
| April    | Exponential            | 5.152       | 11.55           | 1.299               | 30.84            | 15.59           | −0.049| 2.625| 2.630| −0.007| 0.99  |
| May      | Spherical              | 2.99        | 36.76           | 0.976               | 7.54             | 11.71           | −0.075| 2.376| 2.769| −0.008| 0.91  |
| June     | Circular               | 6.17        | 59.31           | 0.919               | 9.42             | 7.258           | 0.0627| 3.186| 3.554| 0.007| 0.97  |
| July     | Stable                 | 6.04        | 233.5           | 1.131               | 2.52             | 7.113           | −0.0984| 2.954| 3.671| −0.005| 0.97  |
| August   | Circular               | 2.33        | 170.2           | 0.835               | 1.35             | 7.034           | −0.1247| 3.333| 4.202| −0.010| 1.05  |
| September| Stable                 | 7.71        | 43.16           | 0.835               | 15.15            | 6.980           | 0.0740| 2.825| 3.193| −0.011| 0.921 |
| October  | Spherical              | 4.03        | 7.375           | 1.171               | 35.33            | 8.899           | −0.0764| 2.455| 2.286| −0.018| 1.04  |
| November | Spherical              | 0.14        | 7.725           | 0.106               | 1.78             | 0.372           | −0.1763| 2.375| 2.536| −0.034| 0.95  |
| December | Stable                 | 3.59        | 7.205           | 0.157               | 33.25            | 1.266           | −0.033 | 2.21 | 2.492| −0.013| 0.86  |
months are generally considered the winter months. This is due to low solar radiation received during these months which is essential for the formation of ozone. Similarly, the higher levels of concentrations were observed during the months of April, May, and June. In these months, the solar radiation level is generally higher, and thus the formation of ozone is more rapid in comparison to the winter months. From the maps, it was found that ozone concentration generally found the maximum in the north-east region of the study area. Part of the coastal area also showed maximum concentrations during the months of October, November, December, and January. Spatial distribution further depends on the wind speed and direction in the region. In future, the precursor concentrations (NOx) distributions and wind speed data will be studied to understand the actual reason for this behaviour.

4. CONCLUSIONS

Kriging is thought to produce the most reliable estimates of values between monitors, and we have successfully applied the method for O3 concentrations within the eastern part of Texas. Kriging maps obtained for each of the twelve months in 2012, indicates that the spatial trends of ozone concentration were not uniform in each case. This may be due to uneven fluctuation in the local climatic conditions from one region to another. Thus, the formation and dispersion process of ozone has also changed unevenly from one region to another. The ozone maps clearly indicate that the concentration values found the maximum in the north-east region of the study area in most of the months. The modeled maps can be used to identify the locations and suggest the high concentration ozone risk zones. Further study is required to identify the reasons for the specific characteristics of ozone distributions in the regions. This will help to take the control measures to reduce the ozone concentrations in risk prone areas for protection of human and animal health.

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