

## Determination of Spatial-Temporal Correlation Structure of Troposphere Ozone Data in Tehran City

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### Abstract

Spatial-temporal modeling of air pollutants, ground-level ozone concentrations in particular, has attracted recent attention because by using spatial-temporal modeling, can analyze, interpolate or predict ozone levels at any location. In this paper we consider daily averages of troposphere ozone over Tehran city. For eliminating the trend of data, a dynamic linear model is used, then some features of correlation structure of de-trended data, such as stationarity, symmetry and separability are considered. Next based on the obtained features, an appropriate model is proposed. This model can be used for future predictions of ozone in Tehran.

**Keywords:** Spatial-temporal process; Dynamic spatial linear model; Stationary; Symmetry; Separability

### Introduction

In recent years there has been a tremendous growth in the statistical models and techniques to analyze environmental processes that are spatially and temporally indexed, such as air pollution data. Troposphere Ozone is a secondary pollutant that results from photochemical reactions involving nitrogen oxides (NO<sub>x</sub>) and volatile organic compounds (VOC's). The rate of ozone production depends on meteorological conditions, primarily sunlight, temperature, along with wind speed and direction. Therefore its levels are difficult to control. A complete description of the chemical processes involving ozone can be found in Seinfeld and Pandis [24]. In 1997, the U.S. Environmental Protection Agency (U.S. EPA) defined the National Ambient Air Quality Standards (NAAQS)

for ozone in terms of the daily 8-hour maximum ozone measurement among the network of monitoring sites covering a given area. The new standard is defined in terms of the 3-year rolling average is less than 80 parts per billion (ppb), (see e.g. [epa.gov/air/criteria.html](http://epa.gov/air/criteria.html)).

Environmental experts and authorities have a special interest in troposphere ozone or ground-level ozone because of its adverse health effects and its impact on certain agricultural crops. Thompson *et al.* [26] represented a comprehensive overview of statistical methods for the statistical adjustment of ground-level ozone. Zhu *et al.* [30] relate ambient ozone and pediatric asthma ER visits in Atlanta using hierarchical regression methods for spatially misaligned data. Wikle [28] provides an overview of hierarchical modeling in environmental science. Cocchi *et al.* [4] followed the approach of Huang and Smith [13] by using a tree based

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partitioning of daily maxima ozone concentrations and assumed these maxima are Weibull distributed. Sahu *et al.* [21] presented a very elegant approach, by using ozone differentials to explain spatial-temporal patterns for ozone in Ohio.

In this paper, we focus on analyzing troposphere ozone for capital city of Iran, Tehran. For statistical analysis of spatial-temporal data, it is often necessary to specify a spatial-temporal correlation structure by covariance function. Ma [19]. The acceptance of some assumptions like stationary, symmetry and separability of spatial-temporal covariance function, would considerably simplify fitting a covariance model to the data. But often they are not applicable, for example with ozone data, separability is not generally a realistic assumption. Nonseparable spatial-temporal covariance models have been proposed by Christakos and Hristopulos [3], Cressie and Huang [7], Christakos [2], De Cesare *et al.* [8,9], Ma [16, 17, 18, 19], Gneiting [11] and Stein [25].

Cressie and Huang [7] based their approach on Fourier transforms. Gneiting [11] proposed another general class of nonseparable, stationary covariance functions for spatial-temporal random processes directly in the spatial-temporal domain. In both these papers the spatial-temporal processes are assumed stationary in time and spatial components. Since the trend of the data arises bias on the covariance function estimation (Cressie [6]), it is necessary to use the de-trended data for fitting a valid covariance function. In this paper, we use a dynamic spatial linear model for modeling trend of ozone data in Tehran city, one of the most polluted cities in Iran. Next, according to the obtained features for correlation structure of this data, a suitable function for covariance structure of the de-trended data is fitted.

In this paper, first, introduce some basic features of the spatial-temporal theory. Next, pertinent exploratory analyses of the data is presented. Our proposed model for modeling trend of spatial-temporal data is described in the Spatial-Temporal Trend section. Then the features of correlation structure of the ozone concentration in Tehran city are specified and a suitable spatial-temporal covariance function is fitted. Finally, Results and Discussion are given.

### Background: Spatial-Temporal Process

Let  $Z(\cdot, \cdot) = \{Z(s, t); s \in R^d, t \in R\}$  denotes a spatial-temporal random field, where  $s$  represents a site in  $d$ -dimensional space and  $t$  represents time. In general, a spatial-temporal random field can be decomposed as

$$Z(s, t) = \mu(s, t) + \delta(s, t); \quad (s, t) \in R^d \times R$$

where  $\mu(s, t)$  represents the mean surface or spatial-temporal trend, also it is corresponding to large scale variations in the process,  $\delta(s, t)$  is a zero mean spatial-temporal correlated error that explains the spatial-temporal small scale variations.

**Definition 1.** The spatial-temporal covariance function is defined as

$$C(s_1, s_2; t_1, t_2) = Cov[\delta(s_1, t_1), \delta(s_2, t_2)]; \\ (s_1, t_1), (s_2, t_2) \in R^d \times R$$

**Definition 2.** The zero mean spatial-temporal process  $\delta(s, t)$  has *stationary* covariance if

$$C(s_1, s_2; t_1, t_2) = C(h; u); \quad (h; u) \in R^d \times R$$

where  $h = s_1 - s_2$  and  $u = t_1 - t_2$ .

**Definition 3.** The spatial-temporal process  $\delta(s, t)$  has a *separable* covariance if there exist purely spatial and purely temporal covariance functions  $C_S$  and  $C_T$ , respectively, such that

$$C(s_1, s_2; t_1, t_2) = C_S(s_1, s_2)C_T(t_1, t_2); \\ (s_1, t_1), (s_2, t_2) \in R^d \times R$$

**Definition 4.** The spatial-temporal process  $\delta(s, t)$  has *fully symmetric* covariance if

$$Cov[\delta(s_1, t_1), \delta(s_2, t_2)] = Cov[\delta(s_1, t_2), \delta(s_2, t_1)]$$

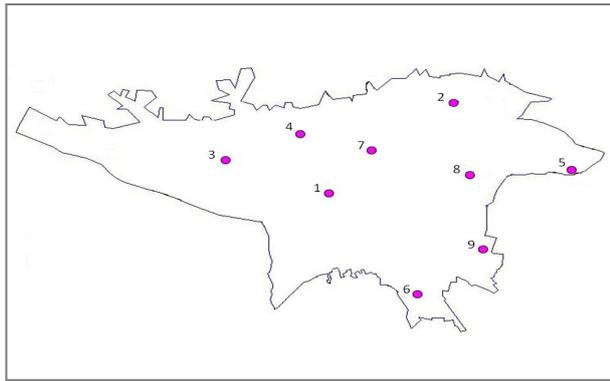
for all spatial-temporal coordinates  $(s_1, t_1)$  and  $(s_2, t_2)$  in  $R^d \times R$  (Gneiting [11]; Stein [25]). Also, a *stationary* spatial-temporal covariance function is *fully symmetric* if

$$C(h, u) = C(h, -u) = C(-h, u) = C(-h, -u); \\ (h, u) \in R^d \times R$$

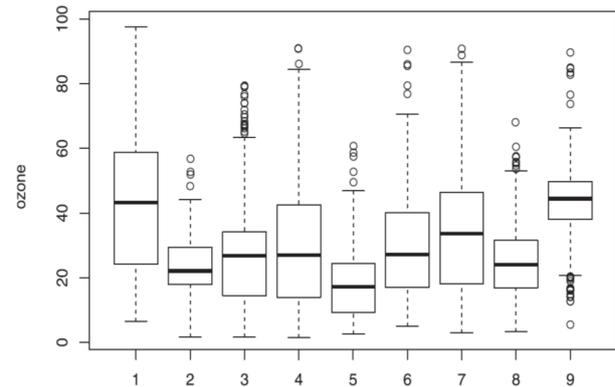
### Exploratory Analysis

In this paper, we have used the daily averages of tropospheric ozone observed during 2009. This data were at scale parts per billion (*ppb*) and have measured at 9 different monitoring stations scattered irregularly in

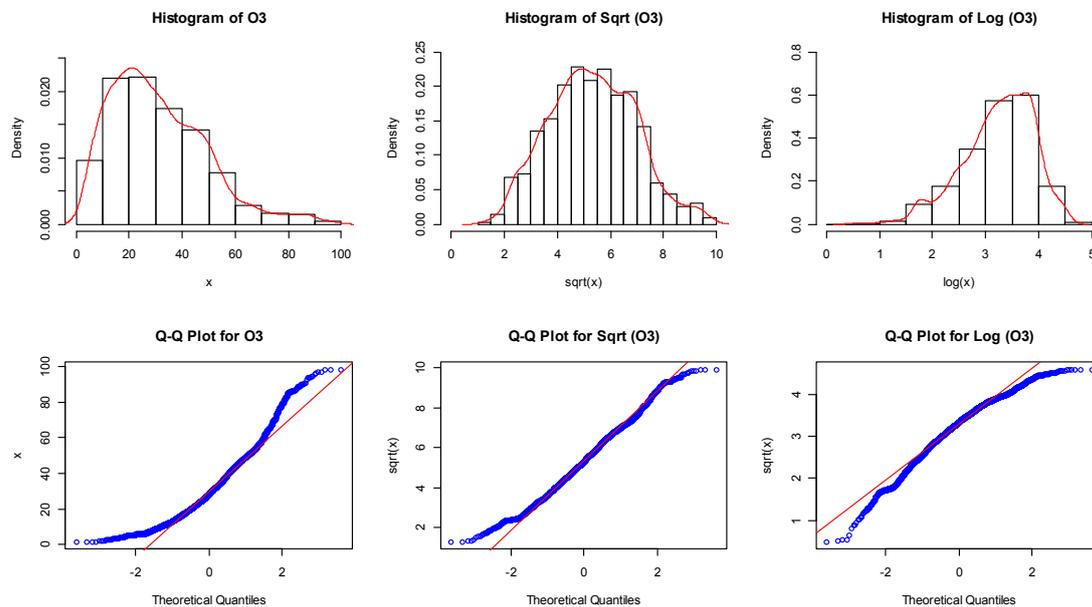
## Determination of Spatial-Temporal Correlation Structure of Troposphere Ozone Data in Tehran City



**Figure 1.** Monitoring sites in Tehran city.



**Figure 2.** Box plot of the data at 9 sites.



**Figure 3.** Histograms and Normal QQ plots for original, square root and logarithmic data.

Tehran city. Fig. 1, shows the geographical locations of these 9 stations over map of Tehran city. Between initial hourly data, some points were missing observation which we imputed them by using of average method, that is, missing observations at each station, is imputed by daily average of all data at the same station.

The box-plot of the data in each station, plotted in Fig. 2, shows considerable spatial variations in this data set. It also shows that the sites 1 and 5 are more and less polluted than others, respectively. Because site 1 is in central area and messy of this city and site 5 is in suburbia out of the city.

For analysis of spatial-temporal data, it is necessary

to consider their normality, stationarity and homogeneity of the variance. Empirical analysis suggested that normality was a reasonable assumption for air pollutant data. For considering normality, the histogram, normal QQ plot and Shapiro-Wilk test for original, square root and logarithm of the data are used. Both of this plots and result of the Shapiro-Wilk test showed that the original and logarithmic data have asymmetric distribution and transformed data by the square root transformation has nearly symmetric distribution (Fig. 3). Also the p-value of Shapiro-Wilk test for the square root of the data is more than 0.05 that approve normality of the transformed data. Therefore we use normal

distribution for the square root of the data.

Since the spatial domain of consideration is large, one could not expect spatial stationarity across this domain. For specifying stationarity in mean and covariance function can be used the H-scatter plots of  $z(s, t)$  versus  $z(s+h, t+u)$  for the original and transformed data that are shown in Fig. 4. In these plots, points are interspersed around bisector line asymmetric. Therefore there is not stationarity for original and transformed data. It can be because of existence of trend in the data.

Cressie and Huang [7], Sahu *et al.* [21] and Huang *et al.* [12] used the plot of standard deviations versus the mean of data (over time) for considering homogeneity of variance. By plotting the standard deviation against the mean of the original data over the 365 time instants analyzed, in Fig. 5.a and over 9 sites in Fig. 5.b, perceive that variance increases as mean increases. Therefore variance of the original data is not homogen. But for the square root of data there is no specific pattern in Fig. 5.c and Fig. 5.d, so the variance of the transformed data is homogen.

**Spatial-Temporal Trend**

For inquiring symmetry and separability of spatial-temporal covariance function, it is needful that eliminate the trend of data. There are variety methods for trend modeling. Cox and Chu [5] used a generalized linear model approach to estimate trends in daily maximum ozone levels. Stroud *et al.* [23] modeled the trend at each time-period as a locally weighted mixture of linear regressions. Huerta *et al.* [14] and Zheng *et al.* [29] applied a dynamic linear model to explain ozone trends. Fuentes *et al.* [10] introduced spectral spatial-temporal models, using covariates that have spatial-temporal dynamic coefficients and applied ambient ozone data provided by U.S. EPA in their article.

Comparing four different models for trend of the ozone data during 2009, Mousavi And Mohammadzadeh [20] proposed a dynamic spatial linear model. In this section, we used the same spatial-temporal model for the transformed data. Then after de-trending the data, in the next section, the symmetry and separability of spatial-temporal structure of the data are considered.

**Dynamic Spatial Linear Model**

Let the  $m$ -dimensional observation vector  $Z(t) = (Z(s_1, t), \dots, Z(s_m, t))$  at time point  $t, t = 1, \dots, n$ , has multivariate Normal distribution  $N_m(\mu(t), V_t)$ .

This model for each  $t$  is defined by observation and evolution equations:

Observation equation:

$$\mu(t) = F_t^T \theta(t) + \epsilon(t), \quad \epsilon(t) \sim N_m(0, V_t),$$

Evolution equation:

$$\theta(t) = G_t \theta(t-1) + \omega(t), \quad \omega(t) \sim N_q(0, W_t)$$

where  $F_t$  is the  $q \times m$  design matrix,  $\theta(t)$  is the  $q \times 1$  state vector,  $\epsilon(t)$  is the observational error vector with  $m \times m$  covariance matrix  $V_t$ ,  $G_t$  is the evolution matrix related to the state vector and  $\omega(t)$  is the evolution error vector with  $q \times q$  covariance matrix  $W_t$ , also  $\epsilon(t)$  and  $\omega(t)$  are independent. This model is

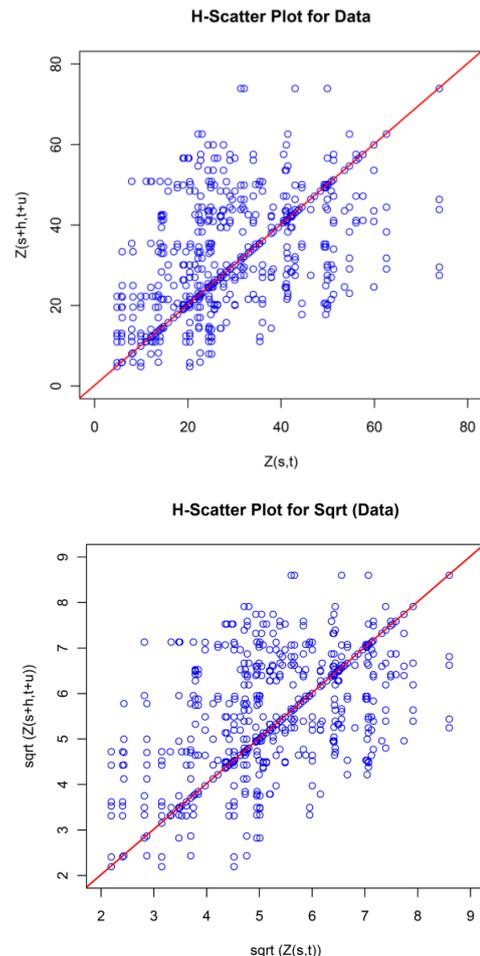
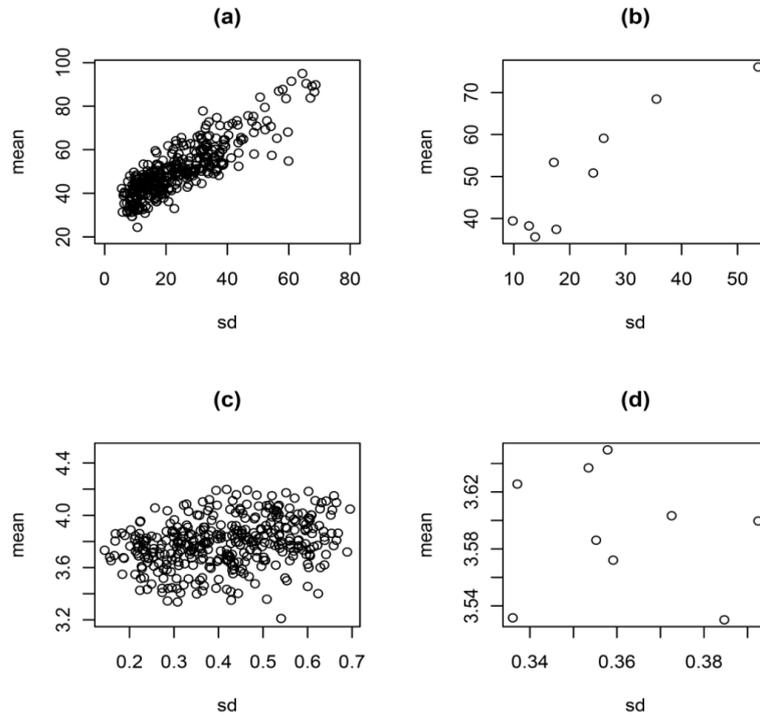


Figure 4. H-Scatter Plots for (a) original data and (b) square root of data.



**Figure 5.** Variation of standard deviation against the mean of the original data (a,c), and square root of data (b,d).

completed with a prior on the initial state vector,  $\theta_0 | \mathbf{D}_0 \sim N(\mathbf{m}_0, \mathbf{C}_0)$ , where  $\mathbf{D}_0$  denotes the initial information set, and  $\mathbf{m}_0$  and  $\mathbf{C}_0$  are known (West and Harrison [27]). Assume that the spatial-temporal observations have cyclical behavior and the state vector can be define as  $\theta(t) = (\theta_1'(t), \theta_2'(t))'$ , where  $q = r + 2k$ ,  $\theta_1'(t)$  is the  $r \times 1$  spatial process and  $2k \times 1$ -dimensional vector  $\theta_2'(t)$  describe cyclical of data. Coefficients of the spatial process characterized with  $\mathbf{X}$  that inclusive length, width, height and other covariates. Corresponding to this partitioning for  $\theta(t)$ , consider design matrix as  $F_t' = (X_t, F_{t_1}, \dots, F_{t_k})$ , where each of the  $F_{t_h}$ ,  $h = 1, \dots, k$ , are  $m \times 2$  matrix that all of elements of the first column are 1 and second column are 0. Therefore for evolution matrix  $\mathbf{G}_t$  can be used a block structure with  $\mathbf{G}_t = \text{blockdiag}(\mathbf{I}_{r \times r}, \mathbf{G}_{t_1}, \dots, \mathbf{G}_{t_k})$ , where each of the blocks  $\mathbf{G}_{t_h}$ ,  $h = 1, \dots, k$ , is a  $2 \times 2$  harmonic matrix of the form

$$\mathbf{G}_{t_h} = \begin{bmatrix} \cos(2\pi h / p) & \sin(2\pi h / p) \\ -\sin(2\pi h / p) & \cos(2\pi h / p) \end{bmatrix}, h = 1, \dots, k$$

For modeling the spatial dependency of the observations, consider the covariance matrix  $V_t = \sigma^2 V_\lambda$ , where  $V_\lambda = \exp(-V / \lambda)$  and elements of  $V$  is determined by a known spatial correlation function, as Matern correlation function. The evolution variance  $W_t$  can be specified either explicitly or through a discount factor  $\alpha \in [0, \infty)$ , which defines  $W_t = \alpha P_t$ , where  $P_t = \text{var}(G_t \theta(t-1) | D_{t-1})$ . A discount factor of  $\alpha = 0$  gives a static model, with the same coefficients for all time periods, whereas  $\alpha \rightarrow \infty$  implies coefficients which are independent over time, i.e. no temporal smoothing at all (Stroud *et al.* [23]).

### Trend of Ozone Data

Let  $Z(s_i, t_j)$  denotes the square root of observed ozone data, at each spatial location  $s_i$ ,  $i = 1, \dots, 9$  and each time  $t_j \in \mathbf{T} = \{1, \dots, 365\}$ . For modeling the trend of the data, we use the available important meteorological variables in monitoring stations, i.e.  $NO$  and  $NO_2$ , which have the most impact on production tropospheric ozone. The number of days, stations and regression coefficients are  $n=365$ ,  $m=9$  and  $q=3$ ,

respectively. We consider

$$F'_t = \begin{bmatrix} 1 & NO(s_1, t) & NO_2(s_1, t) \\ \vdots & \vdots & \vdots \\ 1 & NO(s_9, t) & NO_2(s_9, t) \end{bmatrix},$$

$$\theta(t) = \begin{bmatrix} \theta_1(t) \\ \theta_2(t) \\ \theta_3(t) \end{bmatrix}, G_t = I_{3 \times 3},$$

where, by noting the evolution equation (4), each member of  $\theta(t)$  is simulated from autoregressive model given by

$$\theta_i(t) = \theta_i(t-1) + \omega_i(t), \quad i = 1, 2, 3.$$

The observation errors are assumed to be Gaussian, with mean  $\mathbf{0}$  and covariance  $V_t = V = \sigma^2 \mathbf{I}$ , where  $\mathbf{I}$  is identity matrix of order 9. Leaving  $\sigma^2$  unknown, we select a Gamma prior:  $\frac{1}{\sigma^2} \sim \text{Gamma}(0.01, 0.01)$  so that its mean is 1 and its variance is large. Since usual selection prior for  $\lambda$  is the Uniform distribution  $U(a, b)$ , where  $a$  and  $b$  are minimum and maximum values of spatial lag, we used  $\lambda \sim U(0.051, 0.315)$  where 0.051 and 0.315 are Transformed numbers by Lambert transformation. To complete the model specification, we choose a diffuse prior for the initial state vector:  $\theta(0) | D_0 \sim N_3(0, 100I)$ , where  $\mathbf{I}$  is a  $3 \times 3$  identity matrix. Next the MCMC algorithm was run for 10000 iterations. After a burn in period of 5000 iterations, the Bayesian estimation of the parameters were obtained as shown in Table 1 and also  $R^2 = 0.94$ .

**Correlation Structure of Ozone Data**

To investigate the spatial-temporal correlation structure of the data, first we used a nonparametric test where proposed by Shao and Li [21] to test for symmetry and separability of spatial-temporal covariance functions (Behshad and Mohammadzadeh [1]). Using this testing for the de-trended data, rejected the assumption of separability and didn't reject the assumption of fully symmetry at 5% level. Also H-scatter plot for the de-trended data appear where there is stationary in spatial-temporal covariance structure of this de-trended data. Therefore we consider a symmetric, nonseparable and stationary spatial-temporal covariance function for the ozone data.

**Fitting Covariance Model**

In this subsection we present some models for symmetric nonseparable spatial-temporal stationary covariance functions that introduced by Cressie and Huang [7] and Gneiting [11]. Using method of Cressie and Huang [7] three covariance functions in  $R^d \times R$  were considered as follows

$$C_1(\mathbf{h}, u) = \frac{\sigma^2 (a|u|+1)}{\left[ (a|u|+1)^2 + b \|\mathbf{h}\|^2 \right]^{\frac{d+1}{2}}}$$

$$C_2(\mathbf{h}, u) = \frac{\sigma^2}{(au^2 + 1)^{\frac{d}{2}}} \exp \left\{ -\frac{b \|\mathbf{h}\|^2}{au^2 + 1} \right\}$$

$$C_3(\mathbf{h}, u) = \sigma^2 \exp \left\{ -au^2 - b \|\mathbf{h}\|^2 - cu^2 \|\mathbf{h}\|^2 \right\}$$

where  $a \geq 0$  and  $b \geq 0$  are the scale parameters of the time and spatial lags, respectively,  $c \geq 0$ , and  $\sigma^2 = C(0, 0)$ . Their approach is novel and powerful but depends on Fourier transform pairs in  $R^d$ . In other words, it is restricted to a comparably small class of functions for which a closed-form solution to the  $d$ -variate Fourier integral is known.

The approach of Cressie and Huang [7] was taken by Gneiting [11], but the aforementioned limitation is avoided and very general classes of valid spatial-temporal covariance models are provided. He applied completely monotone functions and positive functions with a completely monotone derivative. Using his method four covariance functions in  $R^d \times R$  were considered as follows

$$C_4(\mathbf{h}, u) = \frac{\sigma^2}{(a|u|^{2\alpha} + 1)^{\beta \frac{d}{2}}} \exp \left\{ -\frac{b \|\mathbf{h}\|^{2\gamma}}{(a|u|^{2\alpha} + 1)^{\beta\gamma}} \right\}$$

$$C_5(\mathbf{h}, u) = \sigma^2 \left[ \frac{c(a|u|^{2\alpha} + 1)}{(a|u|^{2\alpha} + c)} \right]^{\frac{d}{2}} \exp \left\{ -\frac{b \|\mathbf{h}\|^{2\gamma} [c(a|u|^{2\alpha} + 1)]^\gamma}{(a|u|^{2\alpha} + c)^\gamma} \right\}$$

$$C_6(\mathbf{h}, u) = \frac{\sigma^2}{(a|u|^{2\alpha} + 1)^{\beta \frac{d}{2}}} \left[ 1 + \frac{b \|\mathbf{h}\|^{2\gamma}}{(a|u|^{2\alpha} + 1)^{\beta\gamma}} \right]^{-\nu}$$

$$C_7(\mathbf{h}, u) = \sigma^2 \left[ \frac{c(a|u|^{2\alpha} + 1)}{(a|u|^{2\alpha} + c)} \right]^{\frac{d}{2}} \left[ 1 + \frac{b \|\mathbf{h}\|^{2\gamma} [c(a|u|^{2\alpha} + 1)]^\gamma}{(a|u|^{2\alpha} + c)^\gamma} \right]^{-\nu}$$

where  $a$  and  $b$  are nonnegative scale parameters of time and spatial lags, respectively and  $0 < c \leq 1$ . The smoothness parameters  $\alpha$  and  $\gamma$  take values in  $(0, 1]$ ,  $\beta$  is spatial-temporal interaction parameter where take values in  $[0, 1]$ ,  $\sigma^2$  is the variance of the spatial-temporal process and  $\nu > 0$ . Kent *et al.* [15] draw our attention on the counterintuitive presence of a dimple in the spatial-temporal covariance surface in certain cases. That is for a fix spatial lag the temporal covariance is not a decreasing function of the temporal lag as one would normally expect. So we should be careful in applications that the dimple does not accrue at relevant lags.

A weighted-least-squares (WLS) method (Cressie [6]) is used to estimate parameters of each of seven covariance models, by minimizing the criterion given by

$$W(\boldsymbol{\theta}) = \left[ \sum_{i,i',u=1}^U \left( \frac{\hat{C}(\mathbf{h}_{i,i'}, u) - C(\mathbf{h}_{i,i'}, u | \boldsymbol{\theta})}{\sigma^2 - C(\mathbf{h}_{i,i'}, u | \boldsymbol{\theta})} \right)^2 \right]^{\frac{1}{2}}$$

over all possible  $\boldsymbol{\theta}$ . Here  $\mathbf{h}_{i,i'}$  is the spatial lag between stations  $i$  and  $i'$ , and  $u$  is the temporal lag.  $\hat{C}(\mathbf{h}_{i,i'}, u)$  is the empirical correlation given by

$$\hat{C}(\mathbf{h}(\ell), u) = \frac{1}{|N(\mathbf{h}(\ell), u)|} \sum_{(i,j,i',j') \in N(\mathbf{h}(\ell), u)} [Z(s_i, t_j) - \bar{Z}][Z(s_{i'}, t_{j'}) - \bar{Z}]$$

where

$$N(\mathbf{h}(\ell), u) = \{(i, i', j, j') : \mathbf{s}_i - \mathbf{s}_{i'} \in Tol(\mathbf{h}(\ell)); |t_j - t_{j'}| = u; i, i' = 1, \dots, m; j, j' = 1, \dots, n\}$$

Here  $Tol(\mathbf{h}(\ell))$  is some specified tolerance region around  $\mathbf{h}(\ell)$ , and  $|N(\mathbf{h}(\ell), u)|$  is the number of distinct elements in  $N(\mathbf{h}(\ell), u)$ ;  $\ell = 1, \dots, L, u = 1, \dots, U$ .

Table 2 displays comparable parameter estimates for the seven models. Based on the smallest WLS value of

$W(\boldsymbol{\theta})$ , model 4 provides the closest fit to spatial-temporal covariance of ozone data in Tehran city, which its three 3D plot is shown in Fig. 6.

## Results and Discussion

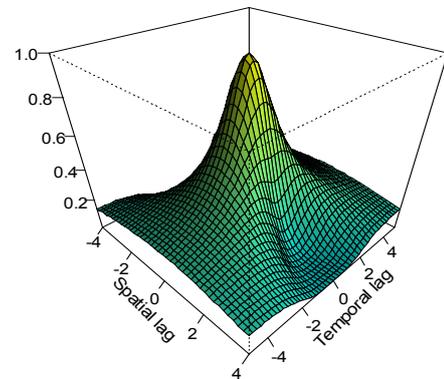
Since ozone concentration data depend to spatial and time locations of observations, we have used a dynamic linear model for modeling trend of these data. After detrending the data, using the test of Shao and Li [22] shows symmetry and nonseparability of spatial-

**Table 1.** Estimation of the model parameters

Parameter	$\theta_1(0)$	$\theta_2(0)$	$\theta_3(0)$	$\sigma$	$\lambda$
Estimated	-0.0295	1.3067	0.1083	0.1584	0.1834

**Table 2.** Estimates of the Parameters of Different Covariance Functions

Covariance model	Parameter						$W(\boldsymbol{\theta})$
	$a$	$b$	$c$	$\alpha$	$\beta$	$\gamma$	
C <sub>1</sub>	0.001	1.252	-	-	-	-	0.3141
C <sub>2</sub>	0.001	0.520	-	-	-	-	0.2463
C <sub>3</sub>	1.001	1.241	0.998	-	-	-	0.2712
C <sub>4</sub>	0.912	0.613	-	0.992	0.989	1	0.1988
C <sub>5</sub>	0.100	0.088	0.998	0.917	0.672	0.962	0.2564
C <sub>6</sub>	0.946	2.955	-	0.045	0.069	1.772	0.016
C <sub>7</sub>	1.053	1.124	0.128	0.459	-	1	0.258



**Figure 6.** Surface of spatial-temporal covariance of Model 4.

temporal correlation structure for the data. The H-scatter plot of  $z(s, t)$  versus  $z(s+h, t+u)$  for de-trended data, shows stationarity in spatial and time. Therefore, correlation structure of these data would be stationary, symmetric and nonseparable. In this paper, seven symmetric and nonseparable spatial-temporal stationary covariance functions are fitted to the data. Among these functions, a Gneiting's model has the smallest WLS value for ozone data in Tehran city, which can be used as a suitable model for correlation structure of this data.

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