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A simple non-separable, non-stationary spatiotemporal model for ozone

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Abstract The past two decades have witnessed an increasing interest in the use of space-time models for a wide range of environmental problems. The fundamental tool used to embody both the temporal and spatial components of the phenomenon in question is the covariance model. The empirical estimation of space-time covariance models can prove highly complex if simplifying assumptions are not employed. For this reason, many studies assume both spatiotemporal stationarity, and the separability of spatial and temporal components. This second assumption is often unrealistic from the empirical point of view. This paper proposes the use of a model in which non-separability arises from temporal non-stationarity. The model is used to analyze tropospheric ozone data from the Emilia-Romagna Region of Italy.

Keywords Spatiotemporal process · Temporal non-stationarity · Non-separability · Deformation · Tropospheric ozone

1 Introduction

Spatiotemporal models arise whenever data are collected across both time and space. Therefore such models have to be analyzed in terms of both their spatial and temporal structures. Until recently, any theories of spatiotemporal processes had always been linked with established theories of spatial statistics and time series analysis. Much of the literature on space-time models is case-specific and tailored to the application in question, the reason for this being the complexity of space-time modelling.

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When a spatiotemporal process is studied, some information about its distribution is usually known, and in general the hypothesis of normality is assumed. When a Gaussian distribution is inappropriate, certain data transformations (like square root, logarithm or Box-Cox transformations) are proposed in order to obtain a more appropriate fitting. Since a Gaussian distribution is fully defined by its first two moments, modelling and analysis will concern the expected value and the covariance structure.

With regard to questions of large-scale variation, the focus of analysis may be placed on the spatiotemporal trend (or expected value); when the study concerns the spatiotemporal connection (small-scale variability), then the focus may be the spatiotemporal covariance (or correlation) function.

Since the spatiotemporal covariance function itself summarizes many aspects of the process, spatiotemporal studies often propose a series of simplifying assumptions about the process in question in order to estimate the spatiotemporal covariance function in the simplest possible way. The most important among them is the separability assumption, as in Rodriguez-Iturbe and Mejia (1974) and De Cesare et al. (1997).

Modelling the levels of tropospheric ozone is important in order to understand and improve air quality in major conurbations. Environmental experts and authorities have a special interest in ozone because of the contribution it makes towards damaging health, the natural environment and materials.

Ground-level ozone has been studied extensively in recent years. The Encyclopedia of Environmetrics features an article on *Ozone* (Cocchi and Trivisano 2002) containing a critical review of the approaches adopted by such studies. Methods for studying extreme events have been proposed by, among others, Cox and Chu (1993) and Smith and Huang (1993), as a method of analyzing daily maximum ozone concentrations. Meanwhile, Guttorp et al. (1994) have developed space-time correlation models designed to spatially interpolate ground-level ozone data. The most recent works also include information on meteorology. Such approaches have been driven by both the desire for more accurate predictive models, and the need to take confounding effects into account when investigating ozone trends and the health effects of ozone. An example is Carroll et al. (1997), who proposed a model designed to evaluate the risk assessment of ozone from emissions, by introducing meteorology into the model.

Our work aims to understand how the commonly proposed assumptions of stationarity and separability may be relaxed, in order to give a more flexible model. We suggest a detailed construction of the model that is constantly checked by data. This model-building approach is developed for the analysis of tropospheric ozone pollution in an area characterized by homogeneous meteorological conditions. Ozone is one of the major pollutants in the area in question, the Emilia-Romagna Region of northern Italy.

The present paper proposes a spatiotemporal model designed to differentiate between diverse mechanisms of non-separability. Section 2 describes the simplifying assumptions that are usually stated for the spatiotemporal covariance function. Section 3 puts forward a spatiotemporal model in which non-separability arises from the non-stationarity of temporal variability. Section 4 describes the ozone data set, while Sect. 5 presents the main results of the analysis. The final section offers a number of conclusions.

2 The simplifying assumptions: stationarity, isotropy and separability

Two kinds of stationarity assumptions are considered within the spatiotemporal context: *strict stationarity* and *second-order stationarity*. A spatiotemporal random field $Z(t, \mathbf{x})$ is considered to be *strictly stationary* within its space-time domain $T \times \mathbf{S}$ if its spatiotemporal law is invariant under translations. This property is difficult to test, since it needs to be demonstrated by considering the family of finite-dimensional distribution functions of the process. Hence a more restrictive property based on moments of the spatiotemporal process is introduced. *Second-order stationarity* involves only the first two moments of the spatiotemporal random field $Z(t, \mathbf{x})$: the mean function is modelled as a constant, and the space-time covariance function is assumed to depend exclusively on the spatial and temporal lags. Strict stationarity only implies second-order stationarity if the first two moments exist, whereas second-order stationarity only implies strict stationarity if the $Z(t, \mathbf{x})$ random field has a Gaussian distribution.

An additional assumption is made for the spatial component of the spatiotemporal correlation function. For many spatiotemporal processes, there is little reason to expect the spatial stationarity of the covariance function. Spatially varying anisotropy can sometimes be modelled through deformations of the geographic coordinates system (Sampson and Guttorp 1992; Damian et al. 2000), where the non-stationary spatial correlation function of a spatiotemporal process is a function of the Euclidean distances between site locations in a bijective transformation of the geographic coordinate system. After an appropriate adjustment has been made to the coordinate system, the correlation structure may be considered isotropic (i.e. it is not dependent on direction but only on distance).

Due to the complexity of modelling spatiotemporal random fields, a *separability* assumption is often made. A spatiotemporal model is deemed *separable* if its space-time covariance function can be written as a product of two functions: one solely a function of space and the other purely a function of time (see for example, Rouhani and Myers 1990 and De Cesare et al. 1997). This simplifying assumption is often introduced more for practical than for empirical reasons, as Cressie and Huang (1999) observed when they wrote: *Separable models are often chosen for convenience rather than for their ability to fit the data well*.

The first attempt at an analysis of non-separability was that of Cressie and Huang (1999). They introduced classes of nonseparable stationary covariance functions to model space-time interactions. They based their approach on Fourier transforms, and used Bochner's Theorem (1955) whereby a continuous function is defined as positive definite if, and only if, it is the Fourier transform of a finite nonnegative measure. Another important contribution to the analysis of non-separable spatiotemporal models was made by Gneiting (2002) who proposed a general class of nonseparable, stationary covariance functions for spatiotemporal random fields directly in the space-time domain (a construction not based on the inversion of a Fourier transformation). By studying the parameter values characterizing this class of covariance functions, one may establish whether separability can be assumed or not. Although these two papers propose different methods of dealing with non-separability, they do share one important feature: the assumption of stationarity in both time and space. A number of statistical tests for separability have been proposed, based on parametric models

(Roberts 2000), likelihood ratio tests and subsampling (Mitchell 2006), or spectral methods (Scaccia and Martin 2005 and Fuentes 2005).

In this paper, we propose a constructive approach designed to introduce a simple method of modelling the case where non-separability arises from temporal non-stationarity. This approach is particularly suitable when the consideration of an annual process may take into account features of seasonal variability other than those encoded in the spatiotemporal trend. This approach is developed by following an environmental application without involving a general class of covariance models, like those of the Cressie and Huang, and Gneiting papers, but providing some insight into a possible generalization.

3 A general spatiotemporal model

A general model for spatiotemporal data $\{Y(t, \mathbf{x})\}$ measured in discrete time $(t = 1, \ldots, T)$ and continuous space $(\mathbf{x} \in \Re^2)$ may be formulated as follows:

$$Y(t, \mathbf{x}) - \mu(t, \mathbf{x}) = Z(t, \mathbf{x}) = W(t, \mathbf{x}) + \epsilon(t, \mathbf{x}); \quad (1)$$

where $\mu(t, \mathbf{x})$ is the spatiotemporal mean field or trend, and the stochastic component is considered, as in Dryden et al. (2005), to be a single term $Z(t, \mathbf{x}) = W(t, \mathbf{x}) + \epsilon(t, \mathbf{x})$, with mean 0 and spatiotemporal correlation function ρ_Z to be further specified. $W(t, \mathbf{x})$ denotes a zero mean smooth Gaussian spatiotemporal underlying process, and $\epsilon(t, \mathbf{x})$ is an independent zero-mean random-error term. The aforesaid model (1) comprises three independent additive components μ, W and ϵ. Because of this independence, model estimation is usually performed component-wise. In some cases the focus is on trend estimation. A broad review of trend formulations given in Kyriakidis and Journel (1999), separates models into two classes, depending on whether the mean component is viewed as deterministic or stochastic. Another review is that of Dimitrakopoulos and Luo (1997), who propose three alternative types of trend: traditional polynomial functions, Fourier expressions and combinations of the two. When covariates are available, a regression-type estimator may effectively represent the trend component (Carroll et al. 1997). When there is no specific focus on trend detection, and no covariate is available, a trend component may be expressed as a simple function of spatial coordinates. Median Polish may be helpful in estimating the components of an additive decomposition in order to separate time and space features (Cressie 1993; Jaynes and Colvin 1997; Bakhsh et al. 2000).

When interest is on the spatiotemporal structure, the process $Z(t, \mathbf{x})$ is analyzed. Separable spatiotemporal models are relatively easy to estimate, but situations requiring nonseparable models are very common. Non-separability may arise from process non-stationarity. In the present paper we have searched for an appropriate transformation f such that the original process Z can be expressed as a function of a separable process Z^*: $Z(t, \mathbf{x}) = f(Z^*(t, \mathbf{x}))$. For a separable process V, the spatiotemporal correlation function becomes the product:

$$\rho_V = \mathrm{Corr}(V(t, \mathbf{x}), V(t', \mathbf{x'})) = \rho_1(|t - t'|)\rho_2(\|\mathbf{x} - \mathbf{x'}\|); \quad (2)$$

with $t, t' = 1, \ldots, T$, and $\mathbf{x}, \mathbf{x'} \in \Re^2$, where ρ_1 and ρ_2 are, respectively, functions of time and space only.

In particular, when non-separability is due to temporal non-stationarity, one possible transformation, leading to $Z(t, \mathbf{x})$ in the spatiotemporal model (1), is as follows:

$$Y(t, \mathbf{x}) - \mu(t, \mathbf{x}) = \sigma_{\langle t \rangle, \mathbf{x}} Z^*(t, \mathbf{x}); \ t = 1, \ldots, T, \ \mathbf{x} \in R^2. \tag{3}$$

In (3), the multiplicative parameter $\sigma_{\langle t \rangle, \mathbf{x}}$ represents the component of non-stationary temporal variability which, when removed, leads to separable space and time correlation components. The subscript $\langle t \rangle$ denotes an empirically derived temporal reference set for this scale parameter, and the subscript $\mathbf{x}$ denotes the spatial coordinates at each site. The $\sigma_{\langle t \rangle, \mathbf{x}}$ parameters can be estimated using different techniques, such as moving average windows in time $\langle t \rangle$ or non parametric methods (Bruno 2004). Let us assume that a large-scale temporal process underlies the separable spatiotemporal process $Z^*(t, \mathbf{x})$ in (3), which can be further modelled as a combination of simpler processes. We thus suggest the decomposition into two components: a time-dependent process, i.e. a single temporal process that applies over the entire spatial domain, denoted by $Z_1(t)$, and a temporally uncorrelated space-time process, $Z_2(t, \mathbf{x})$, which is suitable when the spatiotemporal process does not radically change with time, i.e. when it displays similar temporal structures across space. Model (3) is thus rewritten as:

$$Y(t, \mathbf{x}) - \mu(t, \mathbf{x}) = \sigma_{\langle t \rangle, \mathbf{x}} (\beta_{\mathbf{x}} Z_1(t) + Z_2(t, \mathbf{x})); \ t = 1, \ldots, T, \ \mathbf{x} \in R^2. \tag{4}$$

where $\beta_{\mathbf{x}}$ is a site-specific coefficient for the process $Z_1(t)$, and the subscript $\mathbf{x}$ emphasizes the fact that this may vary from site to site.

A further complication that frequently arises is that of anisotropy, which can be removed by deformation analysis (Sampson and Guttorp 1992; Damian et al. 2000). Deformation may be applied to $Z^*(t, \mathbf{x})$, when the spatiotemporal model considered is (3), or to $Z_2(t, \mathbf{x})$, when model (4) is assumed. In both cases, since the final spatiotemporal model has a spatiotemporal correlation function which is separable between temporal and spatial components, the final spatiotemporal correlation function for a general process V is expressed as follows:

$$\rho_{(d)V} = \mathrm{Corr}(V(t, \mathbf{x}), V(t', \mathbf{x}')) = \rho_1(|t - t'|)\rho_2(\|h(\mathbf{x}) - h(\mathbf{x}')\|); \tag{5}$$

where $t, t' = 1, \ldots, T$, and $\mathbf{x}, \mathbf{x}' \in \Re^2$ and the function h is assumed to be smooth and bijective. The subscript d indicates that deformation analysis has been taken into consideration.

The detailed specification of the spatiotemporal model depends on the specific focus of analysis. In some cases, $Z^*(t, \mathbf{x})$ is the target process, in other applications may be broken down into a number of simpler processes, as in (4). In this latter case, the spatiotemporal correlation function under analysis is ρ_{Z_2} of type (2) or $\rho_{(d)Z_2}$ of type (5).

Empirical spatiotemporal correlations are plotted against distances (spatial lags) and are then fitted by using specific theoretical models (exponential, spherical, Gaussian, etc...). When we consider process $Z(t, \mathbf{x})$, spatiotemporal correlations are interpolated conditional on time, whereas when $Z^*(t, \mathbf{x})$ or $Z_2(t', \mathbf{x}')$ are analyzed, the temporal component of the spatiotemporal correlations in question is already isolated.

4 Application to tropospheric ozone

4.1 Description of the data set

The data set consists of tropospheric ozone measurements from 31 monitoring stations situated throughout the Emilia-Romagna Region of Italy (see Fig. 1). The area is located in the northern part of Italy, and is bounded to the east by the Adriatic Sea, and to the south and west by the Apennines Mountains. The area is quite small, with monitoring sites spread out over a distance of some 280 kms from northwest to southeast. Ozone concentrations are measured on a daily time scale, expressed in terms of daily maximum 8-h moving averages computed from hourly ozone concentration data recorded in micrograms per cubic meter, $\mu g/m^3$, over a five-year period (between 1998 and 2002). Eight-h moving average time series are suggested both by European Community Law (1996) and by U.S. EPA air quality standards (EPA 1998).

The stations shown in Fig. 1 are spaced out unequally: most of them are situated along the main trunk route (the Via Emilia) that crosses the entire region.

In Sect. 3, the proposed model was written without any reference to an empirical data; in this Section, on the other hand, we construct the final model according to empirical evidence, following the same order indicated in Sect. 3. The next sub-section

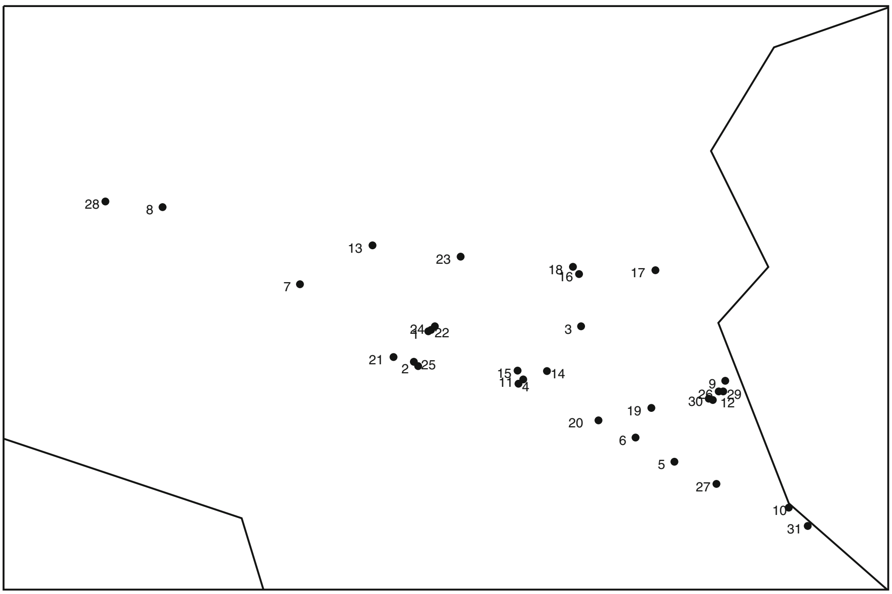

Fig. 1 Map of the monitoring sites in the Emilia-Romagna Region (Italy)

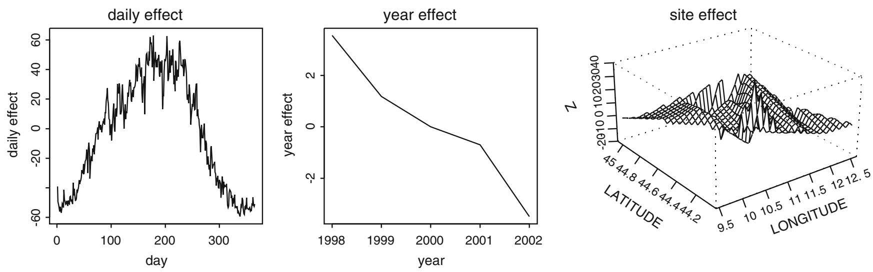

Fig. 2 Trend decomposition by means of Median polish

describes the estimation of trend and seasonal effects, while Subsect. 4.3 gives the reasons for the adoption of the final spatiotemporal model.

4.2 Beginning construction of our model: trend and seasonal effects

The spatiotemporal model is constructed along the same lines as those described in the previous section. The trend component $\mu(t, \mathbf{x})$ in (1) is estimated using a Median Polish algorithm, and includes a seasonal effect (i.e. the annual ozone cycle is very important), a yearly effect and a spatial effect.

The left-hand panel in Fig. 2 shows a seasonal cycle throughout the year, with higher values during the summer days, and lower values during colder winter days. The central panel in Fig. 2 shows the year effect. It seems to be decreasing over the five-year period considered, albeit only slightly. The spatial effect represented in the right-hand panel underlies higher values recorded in the central area of the region. This spatial area corresponds to the largest city in the entire area, Bologna, where ozone pollution can be considered to be higher than in other parts of the region.

4.3 A general spatiotemporal model applied to de-trended data

In this subsection, the decreasing behavior of the spatiotemporal correlation function is modelled exponentially, since this model revealed the best fit, both conditionally on time and after removing the temporal component. An exponential spatial correlation function (see, for instance, Diggle et al. 1998) for $\widehat{Z}(t, \mathbf{x})$ may be expressed as a function of spatial lag $\mathbf{d}$ in (2) for each temporal lag l, as follows:

$$\rho_Z(l, \mathbf{d}) = C(l, \mathbf{d}) = \alpha_l \cdot \exp(-\mathbf{d}/\theta_l). \quad (6)$$

for $l = 0, \ldots, 3$, a range commonly assumed in environmental applications, since temporal correlations fall near to zero beyond 3 days. Figure 3 shows the plots of empirical spatiotemporal correlations ρ_Z against distances (by considering a re-scaled coordinate system with latitude and longitude of between 0 and 1) for the estimated de-trended process $\widehat{Z}(t, \mathbf{x})$ of (1) for temporal lags ranging from 0 to 3.

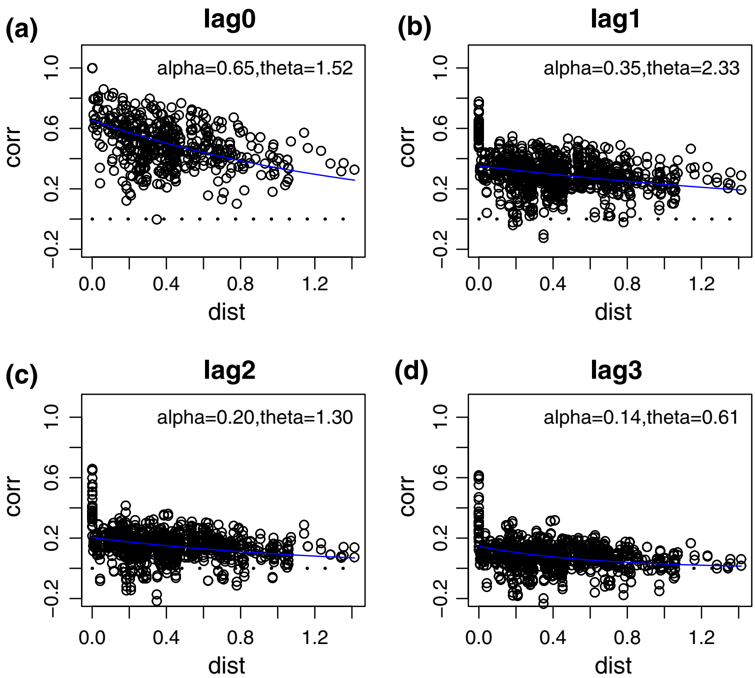

Fig. 3 Spatiotemporal correlations for the process $Z(t, \mathbf{x})$

In a *pure* spatial correlation context, $1 - \alpha$ denotes the *nugget effect* representing micro-scale variations and/or measurement errors. It expresses the discontinuity at the origin, while θ denotes a scale factor.

In the empirical context in question, each panel in Fig. 3 shows the least squares estimates for the two coefficients, α_l and θ_l in (6) for $l = 0, \ldots, 3$. For example, for temporal lag 0 (Figure 3a), the spatial correlations are widely scattered around a fitted exponential model with a nugget effect of approximately 0.35. For each lag, the spatial correlation decreases with spatial distance, but does not reach zero within the region of observations. This is due to the small size of the geographical area in question, and to the uniform influence of meteorology throughout the area. From another point of view, when comparing the panels in Fig. 3 in terms of temporal lag, the spatio-temporal correlations tend, as expected, to decrease with distance and temporal lag. At temporal lag 3, spatiotemporal correlations are very close to zero. The persistence of correlations up to temporal lag 3 could be explained by meteorological conditions producing similar levels for several days.

The de-trended series estimated, $\widehat{Z}(t, \mathbf{x})$, for four sites are plotted in Fig. 4 to highlight the presence of temporal variability that changes seasonally. In fact, this Figure reveals a higher variability on certain specific days of the year, in a plot which is repeated for each year during the period in question.

The temporal non-stationarity in scale (as shown in Fig. 4) suggests the adoption of model (3). The estimation of the $\sigma_{\langle t \rangle, \mathbf{x}}$ component is performed by using a 30-day

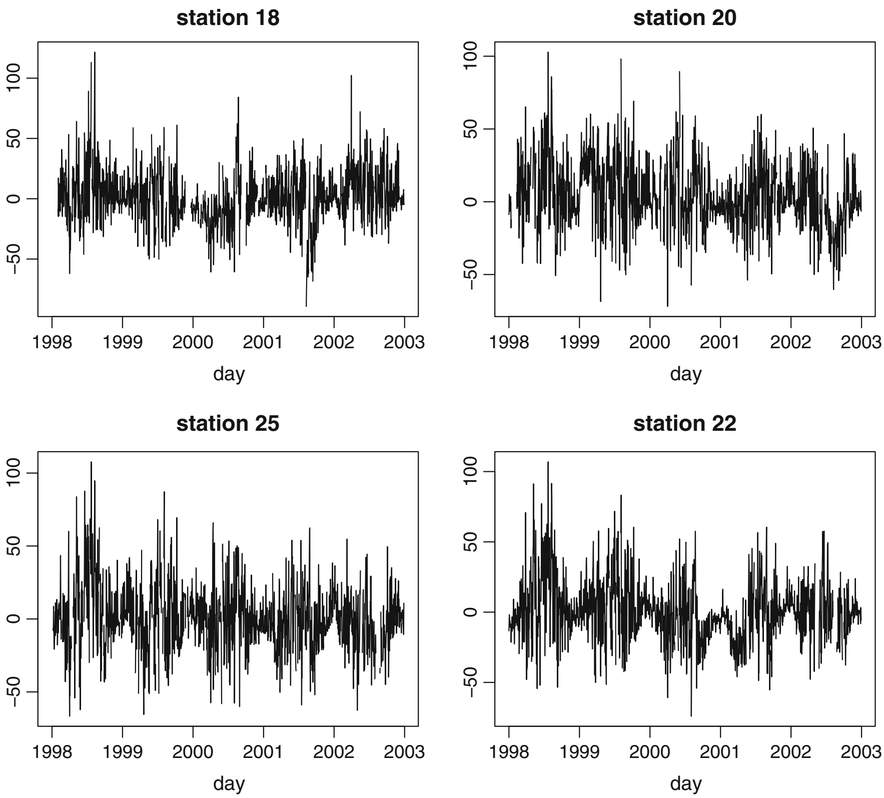

Fig. 4 Ozone time series (after removing the trend)

moving window. In Bruno (2004), non-parametric techniques are also used, although the differences between the results given by the two methods are not substantial.

We can now check whether the introduction of $\sigma_{\langle t \rangle, \mathbf{x}}$ in the model estimation produces a separable spatiotemporal process $Z^*(t, \mathbf{x})$, by assessing whether all sites are characterized by similar temporal structures. Figure 5 depicts AR1 and MA1 coefficients of ARMA(1,1) models fitted at all sites (the magnitude and orientation of the arrow originating from each spatial point corresponds to AR (x-coord) and MA (y-coord) components). The AR1 coefficients average is 0.49, while the MA1 coefficients average is 0.93; the small variability of the coefficients over space indicates the spatial invariance of the structure in time. This lends support to the proposal for a unique time series model for all sites.

The empirical check of the invariance of the temporal correlation structure in space suggests a spatiotemporal process $Z^*(t, \mathbf{x})$ as in (3), with separable spatiotemporal correlation function (2); the temporal correlation ρ_1 shall be expressed as the ARMA(1,1) autocorrelation function found to be suitable for the case in hand, whereas for the spatial correlation function ρ_2 we retain the exponential model (6).

By taking the temporal varying scale parameter $\widehat{\sigma}_{\langle t \rangle, \mathbf{x}}$ into account, process $\widehat{Z}(t, \mathbf{x})$ has been transformed into $\widehat{Z}^*(t, \mathbf{x})$, the empirical spatiotemporal correlations of which

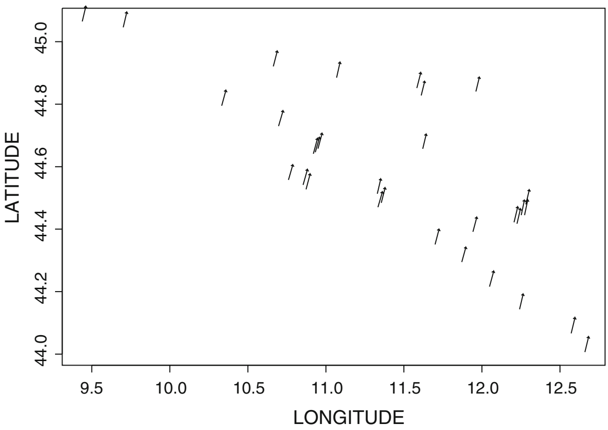

Fig. 5 Relative magnitude of site-specific AR and MA coefficients for all sites

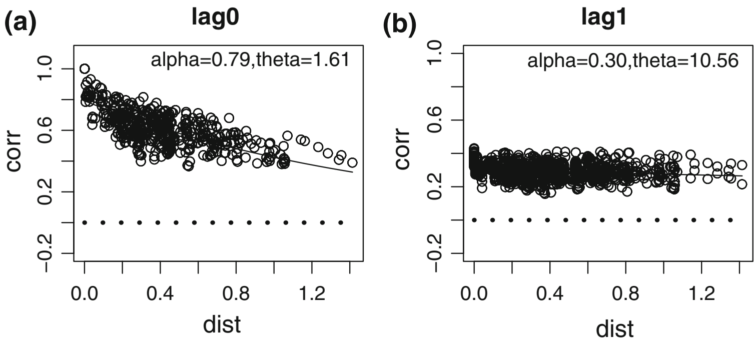

Fig. 6 Spatiotemporal correlations for process $Z^*(t, \mathbf{x})$

are shown in Fig. 6 for temporal lags 0 and 1. A reduction in the scatter of spatial correlations is observed, compared with Fig. 3, even if such correlations remain high regardless of the distances in question.

Moreover, the α and θ least squares estimates of the type (6) exponential correlogram for process $Z^*(t, \mathbf{x})$ are also shown in Fig. 6.

4.4 From Z^* process to Z_1 and Z_2 decomposition

When the spatiotemporal correlations of $\widehat{Z}^*(t, \mathbf{x})$ remain high even for large distances (Figure 6), and the time series have similar structures at each site, we can consider model (4) as appropriate. Indeed, it comprises a large-scale process $Z_1(t)$ and a temporally uncorrelated space-time process $Z_2(t, \mathbf{x})$. The inclusion of the $Z_1(t)$ component in model (4) may mean that the permanence of ozone in the air is spatially uniform. This stands to reason since we know that ozone is a pollutant that behaves stably in air, and has a wide spatial range.

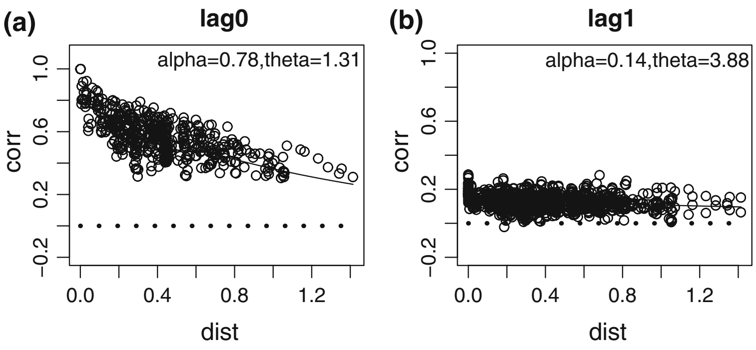

Fig. 7 Spatiotemporal correlations for process $Z_2(t, \mathbf{x})$

In (4) the process $Z^*(t, \mathbf{x})$ is decomposed as the sum of $\beta_{\mathbf{x}} Z_1(t)$ and $Z_2(t, \mathbf{x})$. We estimate the process $Z_1(t)$ using a principal component analysis in which the first principal component incorporating the largest proportion of variability are represented. We also report least squares estimates for α and θ parameters of (6). After estimating the large-scale process, the $Z_2(t, \mathbf{x})$ process is directly estimated, and as such it shall be the focal point of the following discussion of spatial structure. Figure 7 shows the spatiotemporal correlations for process $Z_2(t, \mathbf{x})$ for temporal lags 0 and 1. It also shows least squares estimates for the α and θ parameters of (6).

Removal of the large scale process $Z_1(t)$ enables us to overcome the presence of correlation for large distances, leading to correlations ρ_{Z_2} tending towards zero within the spatial range of the data (see Fig. 7 panel b).

4.5 Anisotropy: deformation analysis

Following the pattern of the Sect. 3, we achieve anisotropy in ρ_{Z_2} as in (5), by adopting the Bayesian model proposed by Damian et al. (2000), in order to estimate heterogeneous spatial covariances by means of a Markov Chain MonteCarlo simulation. Figure 8 shows the posterior mean deformation: it is characterized mainly by compression, meaning high spatial correlation along the northwest-southeast axis traced by the principal road running through the region, especially in the northwest.

Finally, Fig. 9 shows the spatial correlations (temporal lag 0) in the original coordinate system and in the new coordinate system for process $Z_2(t, \mathbf{x})$, respectively, which is designed to evaluate the deformation.

This Figure shows the effect of anisotropy; if we compare panels (a) and (b) one can see that the removal of anisotropy leads to spatial correlations with a smaller dispersion around the fitted exponential correlogram.

4.6 Discussion of the final model

In this Section we are going to justify, and comment on, the proposed final model. Table 1 lists all estimated coefficients of the exponential correlation model (6) and their standard errors (in brackets), together with standard errors of residuals.

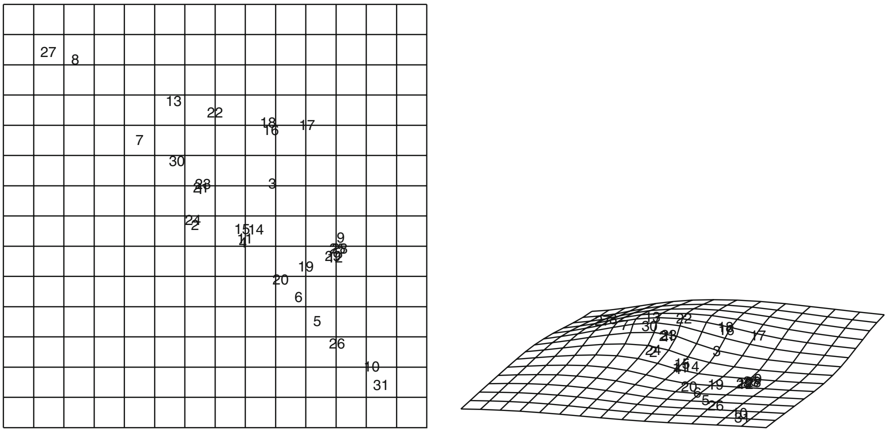

Fig. 8 Original grid coordinates and spatial deformation grid for process $Z_2(t, \mathbf{x})$

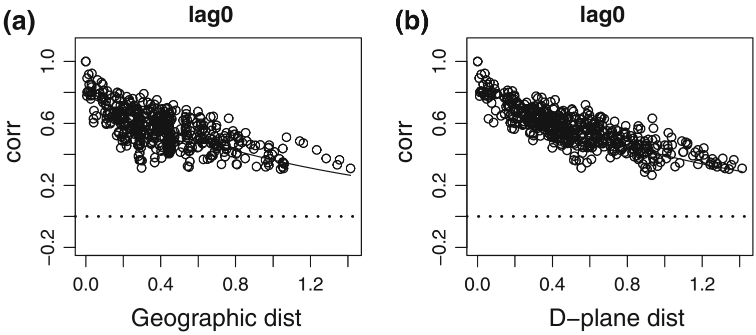

Fig. 9 Spatial correlations for process $Z_2(t, \mathbf{x})$ by original coordinates (panel a) and by coordinates after deformation analysis (panel b)

Parameter α has a spatiotemporal meaning, since it summarizes both the spatial discontinuity at the origin and the temporal correlation structure. The assessment of the nugget effect is a way of comparing models. In the case of process $Z^*(t, \mathbf{x})$ (obtained after removing the non-stationary scale effect $\sigma_{\langle t \rangle, \mathbf{x}}$), the nugget effect is approximately 0.21, which is smaller than the 0.35 estimated for the unscaled process $Z(t, \mathbf{x})$. The nugget has similar values for both $Z^*(t, \mathbf{x})$ and $Z_2(t, \mathbf{x})$. In the case of similar nugget values, differences only derive from the $\widehat{\theta}$ parameters, which represent the correlation distance. By applying deformation analysis to $Z_2(t, \mathbf{x})$, the nugget effect is reduced from 0.22 to 0.13 and the residuals are the smallest. Before deformation analysis was carried out, the spatial correlations of process $Z_2(t, \mathbf{x})$ remain high even for large distances and $\widehat{\theta}$ is higher than 1; whereas when the model has been completed by deformation, spatial correlations tend to zero in correspondence to high distances, and $\widehat{\theta}$ is lower than 1, meaning that there is no longer any spatial correlation beyond the range of the spatial domain in question.

The comparison of Fig. 3 (panel a) with Fig. 9 (panel b) constitutes a graphical representation of Table 1. Both comparisons witness the evaluation of the model effect.

Table 1 Summary of parameter estimates (standard error within brackets) in the exponential correlation function for all processes

Process	Parameter estimate	Lag0	Lag1
$Z(t, \mathbf{x})$	$\widehat{\alpha}$	0.66 (0.01)	0.37 (0.01)
	$\widehat{\theta}$	1.65 (0.10)	2.33 (0.23)
	s.e. of residuals	0.147	0.111
$Z^*(t, \mathbf{x})$	$\widehat{\alpha}$	0.79 (0.01)	0.30 (<0.01)
	$\widehat{\theta}$	1.61 (0.05)	10.56 (1.54)
	s.e. of residuals	0.090	0.046
$Z_2(t, \mathbf{x})$	$\widehat{\alpha}$	0.78 (0.01)	0.14 (<0.01)
	$\widehat{\theta}$	1.31 (0.04)	3.88 (0.48)
	s.e. of residuals	0.098	0.044
$Z_2(t, \mathbf{x})$	$\widehat{\alpha}$	0.87 (<0.01)	
(deformation)	$\widehat{\theta}$	0.83 (0.03)	
	s.e. of residuals	0.076	

5 Conclusions

The aim of the present paper is to develop a strategy for modelling spatiotemporal processes based on specification of their spatiotemporal correlation function. Such an approach is appropriate when the removal of non-stationarity in temporal variability leads to a separable spatiotemporal correlation function. In other words, it suits those situations where non-separability is due to temporal non-stationarity, since the variance of the series depends on time.

As an empirical result of our work, tropospheric ozone displays the following behaviors: a high spatial correlation structure, similar temporal structures along space and strong persistence as temporal lags increase, as shown also in Fassó and Negri (2002a,b). Such observations tend to agree with the findings of other studies conducted in the region on fine particulate (Cocchi et al. 2006; Fassó et al. 2007) and as such confirm that the area may be perceived, for both geographical, meteorological and anthropic reasons, as a part of a unique metropolitan area covering the entire Po Valley.

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