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Stochastic modeling of atmospheric pollution: a spatial time-series framework. Part I: methodology

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Abstract

A geostatistical framework for joint spatiotemporal modeling of atmospheric pollution is presented. The spatiotemporal distribution of concentration levels is modeled as a joint realization of a collection of spatially correlated time series. Parametric temporal trend models, associated with long-term pollution variability are established from concentration profiles at monitoring stations. Such parameters, e.g., amplitude of seasonal variation, are then regionalized in space for determining trend models at any unmonitored location. The resulting spatiotemporal residual field, associated with short-term pollution variability, is also modeled as a collection of spatially correlated residual time series. Stochastic conditional simulation is proposed for generating alternative realizations of the concentration spatiotemporal distribution, which identify concentration measurements available at monitoring stations. Simulated realizations also reproduce the histogram of the sample data, and a model of their spatiotemporal correlation. Such alternative concentration fields can be used for risk analysis studies.

Keywords: Chemical deposition; Stochastic modeling; Monte Carlo simulation

1. Introduction

Atmospheric pollution levels exhibit complex spatiotemporal variability over a wide range of spatial and temporal scales with adverse effects on the environment. Often, spatiotemporal variability cannot be accurately represented via physically based (mechanistic) models, e.g., mathematical models of diffusion and transport of pollutants, due to insufficient knowledge of input parameters. Studying space–time patterns of atmospheric pollution and modeling associated uncertainty is therefore important for understanding the origin and effects of atmospheric pollution on terrestrial ecosystems (NRC, 1986).

Stochastic models provide a framework for routing uncertainty into predictions, and are being increasingly used for analyzing atmospheric pollution levels. Christakos (1992) discusses in detail the use of such models in spatiotemporal analysis; a recent review of geostatistical space–time models can be found in Kyriakidis and Journel (1999). Early approaches to space–time modeling treated time simply as an additional dimension, and traditional geostatistical techniques were applied to the $(2D + 1)$ space–time domain (Eynon and Switzer, 1983). Temporal variability was modeled as an additive component specific to the time direction, e.g., zonal anisotropy (Bilonick, 1985), or as a multiplicative component through separable spatiotemporal covariance models (Christakos, 1992). Mixture models involving both zonal and separable components of variability were also considered (Bilonick, 1988; Haas, 1995). The framework of intrinsic random functions of order K, originally proposed in a purely spatial context (Matheron, 1973) was modified to incorporate a temporal trend by Seguret (1989), and was later adopted in a joint space–time setting by Rouhani and Hall (1989), and Christakos (1992); a recent

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application in the context of sulpfate deposition can be found in Vyas and Christakos (1997).

Time, however, is not just an additional dimension. There are fundamental differences between time and space, such as the non-reversibility of time and the lack of notion of past–present–future in space (Rouhani and Myers, 1990). Approaches involving vectors of time series exploit the typically better-informed time domain, but are limited to predictions at the monitoring station locations (Rouhani and Wackernagel, 1990; Rouhani et al., 1992). Such limitation hinders all important procedures of spatiotemporal mapping. Recently, time-series approaches have been generalized to a continuous spatial domain (Oehlert, 1993), and maps of process levels can be constructed recursively using Kalman filtering (Huang and Cressie, 1996; Wikle and Cressie, 1997).

In this paper, stochastic simulation, i.e., the procedure of generating alternative concentration realizations over the space–time domain of interest (Journel, 1989) is proposed for assessing joint space–time uncertainty. Such simulated concentration realizations reproduce a given spatiotemporal covariance model, and identify measured concentration values at monitoring stations (conditional simulation). Joint spatiotemporal uncertainty is characterized by the probability that concentration levels at a set of locations in space over several time instants be jointly greater than a specified threshold (Haas, 1998). In atmospheric pollution, stochastic simulation can be used to quantify uncertainty associated with a path, along which aerosol values, for example, jointly exceed a regulatory threshold. This task calls for multiple representations of the spatiotemporal process in order to determine how often that path of connected high values appears as existing.

A methodology for stochastic spatiotemporal modeling of atmospheric pollution is developed in this work. Observed concentration levels in space and time are modeled as a joint realization of a collection of space-indexed time series, one for each spatial location. Time-series model parameters are spatially varying, thus capturing space–time interactions. Uncertainty regarding concentration levels at unsampled locations is modeled simultaneously in space and time through stochastic simulation. In Section 2, the modeling framework of spatial time series is presented, and in Section 3, the implications of the proposed methodology for modeling atmospheric pollution, and spatiotemporal phenomena in general, are briefly discussed.

2. A spatial time-series framework

Chemical deposition levels often exhibit clearly interpretable patterns of temporal variability. Seasonal and annual patterns in acid deposition, for example, can be linked to precipitation patterns or seasonality of emissions. In addition, deposition levels are typically measured at spatially coarse monitoring networks during long periods of time. This results into time-rich/space-poor data sets composed of a few tens of stations, each with long time series. It thus makes sense to first capitalize on the time domain, establishing parametric temporal trend models which account for clearly interpretable patterns of temporal variability, and then study any dependence of these model parameters in space.

In the proposed methodology, the spatiotemporal process of interest is modeled as a collection of spatially correlated time series, $\{Z(\mathbf{u},t),\ t\in T\}$, one per location $\mathbf{u}\in D$; here $\mathbf{u}=(x,y)$ denotes the 2D spatial coordinate vector, D denotes the study area, and T the time span of interest. That spatiotemporal process is decomposed into

$$Z(\mathbf{u},t)=M(\mathbf{u},t)+R(\mathbf{u},t)\quad \forall \mathbf{u}\in D\ \ \forall t\in T, \tag{1}$$

where $M(\mathbf{u},t)$ is a stochastic space–time component modeling some "average" smooth variability of the spatiotemporal process $Z(\mathbf{u},t)$, and $R(\mathbf{u},t)$ is a stationary residual component, independent of $M(\mathbf{u},t)$, modeling the higher frequency fluctuations around that trend in both space and time.

The trend component typically models long-term temporal patterns, such as process variability attributed to climatic factors. Other components of variability, e.g., those linked to local weather patterns, are typically accounted for by the stochastic residual process. It should be stressed that the dichotomy of relation (1) is a (subjective) modeling decision: there is no "true" temporal trend component, since there are no trend data. All trends are models resulting from the specific algorithm used to determine them, hence, all trend values instead of being estimated values are actually algorithm-specific model outcomes. The resulting residual component is thus a collective term for all components of variability that are not included in the trend model (Thiébaux, 1997).

Temporal profiles of chemical deposition are not stationary in space, i.e., they vary from one location to another, especially over large (continental) scales. Sulfate deposition in Scandinavia, for example, does not exhibit the same pattern of temporal variability as that in southern Europe, mainly due to the influence of local emission patterns and climatic factors. In addition, spatially varying weather conditions lead to different patterns of temporal variability in regions nearby the ocean and in orographically isolated areas. It is therefore critical to consider spatially non-stationary patterns of temporal variability in the modeling procedure.

In this paper, local parametric models for the temporal trend and residual concentration are first established at the monitoring stations. The spatial distribution of each parameter is then characterized probabilistically via a random function (RF) or random field model, i.e.,

a collection of spatially correlated random variables (RVs). These parameter RFs are in turn specified by their statistics, e.g., histograms and variograms. Such statistics are also parametrized: the variogram of each parameter RF, for example, is parametrized in terms of its range and sill value; these latter parameters are termed hyperparameters in the Bayesian literature (Gelman et al., 1995). The final objective is to determine the posterior distribution, i.e., a model of uncertainty given all available information, of the unknown parameter values defining the temporal trend and residual components at any unmonitored location. In lieu of analytical derivations or numerical integrations typically performed in Bayesian analysis, conditional simulation is adopted for arriving at such posterior parameter distributions. Similar approaches include the family of hierarchical Bayesian models, see for example Wikle and Cressie (1997). The additional inference effort incurred from considering spatially varying parameters is compensated by a more realistic representation of the spatiotemporal process variability.

2.1. Station-specific temporal trend models

The first task is to identify interpretable temporal trend forms at each monitoring station location $\mathbf{u}_\alpha$, such as cyclicity of clear period, e.g., annual cycle, or long-term trends linked to emission control policies. This can be done using time or frequency domain algorithms for signal processing (Chatfield, 1996).

The sample concentration profile $\{z(\mathbf{u}_\alpha, t_i),\ i \in T_\alpha\}$ at each station location $\mathbf{u}_\alpha$ is regarded as a realization of a random process $\{Z(\mathbf{u}_\alpha, t_i),\ i \in T_\alpha\}$, where T_α is the time span of measurements at $\mathbf{u}_\alpha$. This random process $\{Z(\mathbf{u}_\alpha, t_i),\ i \in T_\alpha\}$ is decomposed as

$$Z(\mathbf{u}_\alpha, t_i) = m(\mathbf{u}_\alpha, t_i) + R(\mathbf{u}_\alpha, t_i), \quad t_i \in T_\alpha, \tag{2}$$

where $\{m(\mathbf{u}_\alpha, t_i),\ t_i \in T_\alpha\}$ is a deterministic temporal trend, and $\{R(\mathbf{u}_\alpha, t_i),\ t_i \in T_\alpha\}$ is a stationary, zero mean, stochastic residual process.

The deterministic trend at each station location $\mathbf{u}_\alpha \in D$ is modeled as the sum of $(K+1)$ basis functions of time $f_k(t)$:

$$m(\mathbf{u}_\alpha, t_i) = \sum_{k=0}^{K} b_k(\mathbf{u}_\alpha) f_k(t_i), \quad t_i \in T_\alpha, \tag{3}$$

where $b_k(\mathbf{u}_\alpha)$ is the coefficient (intensity) associated with the kth function $f_k(t_i)$, with $f_0(t_i) = 1$ by convention.

Each basis function $f_k(t)$ is independent of the spatial location $\mathbf{u}$, and should ideally have a physical interpretation pertinent to the entire study region. In other words, $f_k(t)$ should correspond to some physically meaningful component of variability, such as a known annual or seasonal periodic component. Periodicities, especially when physically interpretable, should be incorporated in the deterministic trend $\{m(\mathbf{u}_\alpha, t_i),\ t \in T_\alpha\}$ as a Fourier series, that is a series of sine and cosine functions (Chatfield, 1996).

The $(K+1)$ column vector of coefficients $[b_\alpha] = [b_k(\mathbf{u}_\alpha),\ k = 0, \ldots, K]'$ is modeled at each station location $\mathbf{u}_\alpha$, independently from one location to another, using multiple regression (note that superscript $'$ denotes transposition). More precisely, the sample concentration data at location $\mathbf{u}_\alpha$ are expressed as

$$[z_\alpha] = [f][b_\alpha] + [r_\alpha],$$

where $[z_\alpha]$ is a $T_\alpha \times 1$ column vector of observations available at location $\mathbf{u}_\alpha$, $[f]$ is a $T_\alpha \times (K+1)$ design matrix, and $[r_\alpha]$ is a $T_\alpha \times 1$ column vector of residuals.

The vector of coefficients $[b_\alpha]$ is expressed as a weighted linear combination of the available data $[z_\alpha]$ as: $[b_\alpha] = [\lambda_\alpha][z_\alpha]$, where $[\lambda_\alpha]$ is a $(K+1) \times T_\alpha$ matrix of weights assigned to each of the T_α concentration data. If the matrix $[f]$ is full rank, i.e., its columns are linearly independent, then the previous system has a unique solution and the resulting vector of weights $[\lambda_\alpha]$ is given by the ordinary least-squares (OLS) solution as $[\lambda_\alpha] = ([f]'[f])^{-1}[f]'$ (Searle, 1971).

Once the vector $[b_\alpha]$ of $(K+1)$ coefficients specific to each station location $\mathbf{u}_\alpha$ is determined, the algorithm-specific temporal trend model $\{m(\mathbf{u}_\alpha, t_i),\ t_i \in T_\alpha\}$ at that location is given by expression (3), and the corresponding residual series are obtained as

$$r(\mathbf{u}_\alpha, t_i) = z(\mathbf{u}_\alpha, t_i) - \sum_{k=0}^{K} b_k(\mathbf{u}_\alpha) f_k(t_i), \quad i = 1, \ldots, T_\alpha. \tag{4}$$

In the proposed approach, the $(K+1)$ station-specific temporal trend coefficients are defined via the algorithm adopted for their construction; these coefficients are treated as precise data. Alternatively, one could use the variance of the OLS estimator as a measure of local random variability, which could be viewed as part of a nugget component when inferring the spatial cross-variograms of the coefficient fields (see next section). This component could be suppressed (filtered) via factorial kriging (Goovaerts, 1997).

In most regional-scale applications, climatic long-term patterns explain different proportions of the process temporal variability from location to location. Such a spatially varying influence of the trend, and consequently of the residual, component is captured by the location-dependent variance of the residual concentration profiles, see Section 2.4.

2.2. Regionalizing temporal trend coefficients

Recall that temporal trend models $\{m(\mathbf{u}_\alpha, t),\ t \in T\}$ are established independently at each station location $\mathbf{u}_\alpha$. The resulting temporal trend model parameter values,

i.e., the entries of the n parameter vectors $[b_\alpha]$, $\alpha = 1, \ldots, n$, one per location $\mathbf{u}_\alpha$, are spatially correlated since they are derived from the same process data $[z_\alpha]$, themselves correlated in space and time. Spatiotemporal interactions between the trend components are accounted for by correlating in space the local trend model parameters (b_k-values). A similar approach was proposed by Oehlert (1993); the methodology developed hereafter extends this approach to a simulation context, accounting for spatial cross-correlation between the temporal trend parameters.

In the proposed approach, a stochastic spatiotemporal trend model $M(\mathbf{u}, t)$ is defined by viewing the set of $(K+1)$ trend coefficients as a joint realization of a set of $(K+1)$ cross-correlated RFs $\{B_k(\mathbf{u}),\ \mathbf{u} \in D\}$, $k = 0, \ldots, K$, i.e.

$$M(\mathbf{u}, t) = \sum_{k=0}^{K} B_k(\mathbf{u}) f_k(t), \quad \forall \mathbf{u} \in D,\ \forall t \in T, \tag{5}$$

where T denotes the constant time span, same over the entire domain D.

One could physically interpret the spatial correlation between temporal trend model parameters (b_k-values). A model of spatial correlation for any two slope b_1-values, for example, could be viewed as a measure of the influence of emission control policies implemented in any two locations to the rate of decrease or increase of chemical deposition at these locations. Similarly, a model of spatial correlation between any two phase values derived from spectral analysis at two monitoring stations could quantify the influence of transport processes on peak trend values at these stations.

In a simulation context, a joint realization of the $(K+1)$ coefficient values $\{b_k^{(s)}(\mathbf{u}),\ \mathbf{u} \in D\}$, $k = 0, \ldots, K$, determines the sth realization of the spatiotemporal trend field $\{m^{(s)}(\mathbf{u}, t),\ \mathbf{u} \in D,\ t \in T\}$ over the space–time domain $D \times T$, as

$$m^{(s)}(\mathbf{u}, t) = \sum_{k=0}^{K} b_k^{(s)}(\mathbf{u}) f_k(t), \quad \forall \mathbf{u} \in D,\ t \in T, \tag{6}$$

where the superscript (s) designates the sth simulated realization.

Such simulation calls for modeling covariance matrix of the vector RF $\{\mathbf{B}(\mathbf{u}),\ \mathbf{u} \in D\}$, where $\mathbf{B}(\mathbf{u}) = [B_k(\mathbf{u}),\ k = 0, \ldots, K]'$. This allows reproduction of any cross-correlation between b_k-coefficients at lag $|\mathbf{h}| = 0$. For example, the negative correlation between the intercept and the linear gradient fields, $B_0(\mathbf{u})$ and $B_1(\mathbf{u})$, which is inherent to any line-fitting procedure, is reproduced by the corresponding simulated realizations.

2.3. Simulation of spatiotemporal trend

Simulation of the spatiotemporal trend reduces to independent simulation of a set of uncorrelated factors, resulting from principal component analysis (PCA) of the $(K+1)$ sets of trend coefficients. PCA amounts to transforming a set of correlated variables, the b_k-coefficients in this case, into uncorrelated (at $|\mathbf{h}| = 0$) x_k-components termed factors, each component being a specific linear combination of the original variables (Wackernagel, 1995). Simulated realizations of the set of $(K+1)$ uncorrelated factors $X_k(\mathbf{u})$ at location $\mathbf{u}$ are generated independently one from another, and the set of $(K+1)$ simulated coefficients of the temporal trend are retrieved from these simulated values.

More specifically, a joint conditional realization $\{b_k^{(s)}(\mathbf{u}_j),\ j = 1, \ldots, N\}$, $k = 0, \ldots, K$, of the $(K+1)$ coefficient RFs $\{B_k(\mathbf{u}),\ \mathbf{u} \in D\}$, $k = 0, \ldots, K$, over N grid nodes discretizing the domain D is generated by: (a) generating a realization $\{x_k^{(s)}(\mathbf{u}_j),\ j = 1, \ldots, N\}$, of the $(K+1)$ factor RFs $\{X_k(\mathbf{u}),\ \mathbf{u} \in D\}$, $k = 0, \ldots, K$, independently one from another; this realization identifies the $n \times (K+1)$ respective sample factor data $\{x_k(\mathbf{u}_\alpha),\ \alpha = 1, \ldots, n\}$, $k = 0, \ldots, K$, and (b) retrieving the simulated value $b_k^{(s)}(\mathbf{u}_j)$ of the kth coefficient at each node $\mathbf{u}_j$ as

$$b_k^{(s)}(\mathbf{u}_j) = \sum_{k'=0}^{K} h_{kk'} x_k^{(s)}(\mathbf{u}_j) s_{B_{k'}} + m_{B_{k'}} \quad \mathbf{u}_j \in D,$$

where $h_{kk'}$ are entries of the matrix $\mathbf{H} = \mathbf{Q}^{-1}$, with $\mathbf{Q}$ being the matrix of eigenvectors associated with the correlation matrix of the $K+1$ coefficients, $m_{B_{k'}}$, $s_{B_{k'}}$ are the mean and standard deviation of the k'th coefficient values.

Any conditional simulation algorithm can be employed for generating the S realizations of each factor RF $\{X_k(\mathbf{u}),\ \mathbf{u} \in D\}$, e.g., Gaussian or direct sequential simulation (Journel, 1994; Deutsch and Journel, 1998). The orthogonality of the principal components at lag $|\mathbf{h}| = 0$, which is ensured by construction and for a multivariate Gaussian RF implies their independence, is assumed to extend to all other separation vectors $\mathbf{h}$, i.e., $\mathrm{Cov}\{X_k(\mathbf{u}), X_{k'}(\mathbf{u}+\mathbf{h})\} \simeq C_{kk'}(\mathbf{h}) = 0$, $\forall k \neq k'$, $\forall \mathbf{h}$. This assumption is valid if the $(K+1)$ variables are intrinsically correlated, i.e., any cross-covariance $C_{kk'}(\mathbf{h})$ is proportional to a single common auto-covariance $C_k(\mathbf{h}) = C(\mathbf{h})$. If this is not the case, then all auto and cross-covariance functions $C_{kk'}(\mathbf{h})$ should be jointly modeled via the linear model of coregionalization (Journel and Huijbregts, 1978; Wackernagel, 1995).

2.4. Location-specific temporal residual models

The spatiotemporal residual process $\{R(\mathbf{u}, t),\ \mathbf{u} \in D,\ t \in T\}$ is modeled as a collection of spatially correlated residual time series (TS), one for each location $\mathbf{u} \in D$. Sample residual profiles $\{r(\mathbf{u}_\alpha, t_i),\ i = 1, \ldots, T_\alpha\}$, $\alpha = 1, \ldots, n$, are first standardized to unit variance by dividing each residual value $r(\mathbf{u}_\alpha, t_i)$ at location $\mathbf{u}_\alpha$ by the standard deviation $s_R(\mathbf{u}_\alpha)$ of the residual profile at that location.

Such a standardization step amounts to defining the residual spatiotemporal field

$$R(\mathbf{u},t) = S_R(\mathbf{u}) \cdot \hat{R}(\mathbf{u},t), \quad \mathbf{u} \in D,\ t \in T \tag{7}$$

with the scale RF $\{S_R(\mathbf{u}),\ \mathbf{u} \in D\}$ having unit mean, and being uncorrelated with the RF $\{\hat{R}(\mathbf{u},t),\ \mathbf{u} \in D,\ t \in T\}, \forall t$.

The spatially varying residual variance value $s^2_R(\mathbf{u})$ at each location $\mathbf{u}$ quantifies the amount of temporal concentration variability, which is accounted for ("explained") by the residual component at $\mathbf{u}$. A low residual variance $s^2_R(\mathbf{u})$ implies that the trend component at location $\mathbf{u}$ provides a close approximation of the actual concentration profile at that location. Conversely, a high residual variance $s^2_R(\mathbf{u})$ implies that most of the concentration temporal variability is modeled ("absorbed") by the residual component.

Next, consider the following decomposition (a model) of the TS $\{\hat{R}(\mathbf{u}_\alpha,t),\ t \in T\}$ into $(L+1)$, zero mean, independent component TS $\{\hat{R}_l(\mathbf{u}_\alpha,t),\ t \in T\}, l = 0,\ldots,L$, as

$$\hat{R}(\mathbf{u}_\alpha,\ t) = \sum_{l=0}^{L} w_l(\mathbf{u}_\alpha)\hat{R}_l(\mathbf{u}_\alpha,t), \quad \alpha \in (n) \tag{8}$$

with $\mathrm{Cov}\{\hat{R}_l(\mathbf{u}_\alpha,t),\ \hat{R}_{l'}(\mathbf{u}_\alpha,t+\tau)\} = \delta_{ll'}C_{\hat{R}_l}(\tau;\ \alpha)$, where $\delta_{ll'}$ is the Kronecker delta, i.e., $\delta_{ll'} = 1$ if $l = l'$, zero if not; $C_{\hat{R}_l}(\tau;\ \alpha)$ denotes the covariance of the lth component TS $\{\hat{R}_l(\mathbf{u}_\alpha,t),\ t \in T\}$, with $\tau = t - t'$ being a temporal lag distance.

The covariance function $C_{\hat{R}}(\tau;\ \alpha)$ of the standardized residual TS $\{\hat{R}_l(\mathbf{u}_\alpha,t),\ t \in T\}$ defined in Eq. (8) is parametrized as

$$C_{\hat{R}}(\tau;\ \boldsymbol{\theta}(\mathbf{u}_\alpha)) = \sum_{l=0}^{L} p_l(\mathbf{u}_\alpha)C_{\hat{R}_l}(\tau;\ q_l(\mathbf{u}_\alpha)),$$

where $p_l(\mathbf{u}_\alpha) = (w_l(\mathbf{u}_\alpha))^2$ and $q_l(\mathbf{u}_\alpha)$ are the positive weight (sill) and range of the lth basic component covariance $C_{\hat{R}_l}(\tau;\ \alpha)$. The sill $p_l(\mathbf{u}_\alpha)$ and range $q_l(\mathbf{u}_\alpha)$ values quantify the amount of temporal variability and the correlation length associated with the lth component TS $\{\hat{R}_l(\mathbf{u}_\alpha,t),\ t \in T\}$. By convention, $q_0(\cdot) = \varepsilon$ is a very small range, which corresponds to a white noise process or nugget effect. These $2(L+1)$ parameters comprise the station-specific parameter vector $\boldsymbol{\theta}(\mathbf{u}_\alpha) = [\{p_l(\mathbf{u}_\alpha), q_l(\mathbf{u}_\alpha)\},\ l = 0,\ldots,L]'$.

For simplicity, all $(L+1)$ basic structures $C_{\hat{R}_l}(\tau;\ q_l(\mathbf{u}_\alpha))$ are assumed here to be of the same type, e.g., spherical or exponential covariance models. The number $(L+1)$ of component TS $\{\hat{R}_l(\mathbf{u}_\alpha,t),\ t \in T\}$ could be also made location-specific; here it is assumed constant. In most practical applications, the temporal correlation of the residual concentration profiles can be modeled by a nugget effect and an exponential structure, the latter corresponding to a first-order auto-regressive process (Chatfield, 1996).

By viewing the set of $(L+1)$ sill p_l-parameters, and the set of $(L+1)$ range q_l-parameters as a joint realization of a set of $2(L+1)$ cross-correlated RFs $\{[P_l(\mathbf{u}), Q_l(\mathbf{u})],\ \mathbf{u} \in D\}, l = 0,\ldots,L$, the standardized spatiotemporal residual field $\hat{R}(\mathbf{u},t)$ is defined as

$$\hat{R}(\mathbf{u},t) = \sum_{l=0}^{L} \sqrt{P_l(\mathbf{u})}\hat{R}_l(\mathbf{u},t), \quad \mathbf{u} \in D,\ t \in T. \tag{9}$$

The location-specific covariance function $C_{\hat{R}}(\tau;\ \boldsymbol{\theta}(\mathbf{u}))$ of the TS $\{\hat{R}(\mathbf{u},t),\ t \in T\}$ is then written as $C_{\hat{R}}(\tau;\ \boldsymbol{\theta}(\mathbf{u})) = \sum_{l=0}^{L} p_l(\mathbf{u})C_{\hat{R}_l}(\tau;\ q_l(\mathbf{u}))$, with $p_l(\mathbf{u}) = (w_l(\mathbf{u}))^2$ now being a realization of the RV $P_l(\mathbf{u})$ defined at location $\mathbf{u}$, and $q_l(\mathbf{u})$ is a realization of the RV $Q_l(\mathbf{u})$ defined at the same location.

In practice, the n temporal covariance models $C_{\hat{R}}(\tau;\ \boldsymbol{\theta}(\mathbf{u}_\alpha))$ of the n standardized residual processes $\{\hat{R}(\mathbf{u}_\alpha,t),\ t \in T\}$ are inferred independently at each station location $\mathbf{u}_\alpha$, and their parameters are regionalized in space accounting for their spatial correlation. This entails regionalization of the $(L+1)$ sill parameters $\{P_l(\mathbf{u}),\ \mathbf{u} \in D\},\ l = 0,\ldots,L$, as well as regionalization of the L-range parameters $\{Q_l(\mathbf{u}),\ \mathbf{u} \in D\},\ l = 1,\ldots,L$, which leads to a model $C_{\hat{R}}(\tau;\ \boldsymbol{\theta}^*(\mathbf{u}))$ for the covariance function of the residual TS at any unmonitored location $\mathbf{u}$. Recall that a nugget effect is characterized by a very small, but constant, range $q_0(\cdot) = \varepsilon$, hence the need to regionalize only L-range parameters.

2.4.1. Simulation of spatiotemporal residual

Simulation of the spatiotemporal residual amounts to generating realizations of a series of 1D processes, the standardized residual process $\{\hat{R}(\mathbf{u},t),\ t \in T\}$ at each location $\mathbf{u}$, using the corresponding covariance model $C_{\hat{R}}(\tau;\ \boldsymbol{\theta}(\mathbf{u}))$. Any stochastic simulation algorithm can be used for simulating these residual profiles, e.g., sequential simulation or simulation via Cholesky decomposition of the covariance matrix (Deutsch and Journel, 1998).

Simulated realizations of the spatiotemporal residual field $\hat{R}(\mathbf{u},\ t)$ exhibit both temporal and spatial correlation. Temporal correlation is imposed via the (location-specific) temporal covariance model $C_{\hat{R}}(\tau;\ \boldsymbol{\theta}^*(\mathbf{u}))$. Spatial correlation is induced via the spatial correlation of the entries of the parameter vector $\boldsymbol{\theta}^*(\mathbf{u})$, i.e., via the covariance models $C_{P_l}(\mathbf{h})$ and $C_{Q_l}(\mathbf{h})$ of the sill and range values of each component TS $\{\hat{R}_l(\mathbf{u},t),\ t \in T\}$. A non-conditional realization $\{\hat{r}^{(s)}(\mathbf{u},t),\ t \in T\}$ of standardized residual profile at any location $\mathbf{u}$, however, cannot be generated independently from a realization $\{\hat{r}^{(s)}(\mathbf{u}',t),\ t \in T\}$ at another location $\mathbf{u}'$, even if the corresponding two parameter vectors $\boldsymbol{\theta}^*(\mathbf{u})$ and $\boldsymbol{\theta}^*(\mathbf{u}')$ are spatially correlated; the reason is that the random numbers used for simulating the two TS (e.g., Chatfield, 1996) at locations $\mathbf{u}$ and $\mathbf{u}'$ should not be drawn independently at these locations. Additional spatial correlation is induced by an unobservable random field of innovations, which is spatially correlated and serially uncorrelated (Wikle and Cressie, 1997; Wilks, 1998). If sequential Gaussian simulation, i.e., autoregression, is used for simulating

standardized residual $\hat{r}$-profiles at each unmonitored location $\mathbf{u}$, such innovation values are spatially colored and temporally white standard normal deviates; their spatial covariance could be identified to that of the sample standardized $\hat{r}$-residuals after the latter are transformed to a Gaussian distribution.

The above innovations field is also used for indirectly conditioning simulated $\hat{r}$-profiles to sample residual $\hat{r}$-data measured at nearby monitoring stations. When simulating a non-conditional realization $\{\hat{r}^{(s)}(\mathbf{u}, t_i),\ i = 1, \ldots, T\}$ of the standardized residual TS $\{\hat{R}(\mathbf{u}, t),\ t \in T\}$ at location $\mathbf{u}$, sample residual $\hat{r}$-data available at nearby monitoring stations are not taken into account; only previously simulated $\hat{r}$-values at $\mathbf{u}$. Consequently, a high simulated residual value $\hat{r}^{(s)}(\mathbf{u}, t_i)$ can be generated next to a low sample residual $\hat{r}(\mathbf{u}_\alpha, t_i)$ measured at the same time instant t_i at an adjacent station location $\mathbf{u}_\alpha$. In the context of sequential Gaussian simulation, indirect conditioning to contemporaneous $\hat{r}$-data can be achieved by conditioning innovation realizations to the normal scores (quantiles of a Gaussian distribution) of the contemporaneous sample $\hat{r}$-data. Such a conditioning step results into simulating high innovation values, and consequently high $\hat{r}$-values, nearby high sample $\hat{r}$-data (Kyriakidis, 1999).

Simulated realizations $\{z^{(s)}(\mathbf{u}, t),\ t \in T\}$ of concentration profiles are finally built by adding the simulated trend $\{m^{(s)}(\mathbf{u}, t),\ t \in T\}$ and residual $\{r^{(s)}(\mathbf{u}, t),\ t \in T\}$ profiles. Since the spatiotemporal trend and residual components are assumed uncorrelated, see definition (1), simulation of the trend field is performed independently from that of the residual field. Often, concentration records $\{z(\mathbf{u}_\alpha, t_i),\ i = 1, \ldots, T_\alpha\}$ at the n monitoring station locations $\mathbf{u}_\alpha$, $\alpha \in (n)$ are not complete. In this case, the missing values of the station profiles are in-filled by stochastic simulation. The set of S alternative, equally probable, realizations $\{z^{(s)}(\mathbf{u}, t), \mathbf{u} \in D, t \in T\}$ provide a model of uncertainty for the unknown process levels in both space and time, which can be used for risk assessment studies or for deciding on additional sampling (Haas, 1998).

3. Discussion

A framework for stochastic spatiotemporal modeling of atmospheric pollution has been presented in this paper. Observed concentration levels are viewed as a joint realization of a collection of spatially correlated time series, thus capitalizing on the typically better informed time domain. The stochastic spatiotemporal field is decomposed into a stochastic trend and a stochastic residual component. Parametric temporal trend models are established at all monitoring stations, independently from one location to another, and their parameters are coregionalized in space. This amounts to mapping the task of simulating a space–time trend field to that of generating realizations of a set of cross-correlated RFs modeling the joint spatial distribution of the temporal trend parameters. Joint conditional simulation of such parameters ensures reproduction of important cross-correlations between them. Simulated realizations of the spatiotemporal residual component are finally generated via a series of TS realizations, whose parameters are correlated in space.

The proposed methodology is a (distributed parameter) spatial time series framework merging modeling approaches typically applied independently in a spatial and temporal context; this comprehensive framework allows a consistent modeling of spatial and temporal variability through stochastic simulation. Concentration profiles obtained from mechanistic model predictions at each location could alternatively be viewed as local trend components (Venkatram, 1988), and the spatiotemporal distribution of the resulting residual profiles could then be modeled along the lines proposed in this work. The set of alternative concentration realizations provides a model of uncertainty regarding the unknown concentration levels in both space and time. Such an uncertainty model could be used in a risk analysis context for studying the impact of emission control policies or for designing monitoring networks.

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