Trends in Rural Sulfur Concentrations

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Abstract

This paper presents an analysis of regional trends in atmospheric concentrations in sulfur dioxide (SO_2) and particulate sulfate (SO_4^{2-}) at rural monitoring sites in the Clean Air Act Status and Trends Monitoring Network (CASTNet) from 1990 to 1999. A two-stage approach is used to estimate regional trends and standard errors in the Midwest and Mid-Atlantic regions of the U.S. In the first stage, a linear regression model is used to estimate site-specific trends in data adjusted for the effects of season and meteorology. In the second stage, kriging methodology based on the maximum likelihood estimation is used to estimate regional trends and standard errors. The method is extended to include a Bayesian analysis to account for the uncertainty in estimating the spatial covariance parameters. Both spatial prediction techniques produced similar results in terms of regional trends and standard errors.

Keywords: CASTNet, regional trends, kriging, Bayesian methods.

1 Introduction

The reliable estimation of temporal trends in pollution is of considerable interest to the scientific community and environmental managers for assessing the effectiveness of emission reductions mandated by the Clean Air Act Amendments (CAAA) of 1990. To determine whether emission reductions have had the intended effect of reducing pollution, airborne concentrations can be statistically modeled to reveal the magnitude and spatial distribution of trends. The identification of long-term trends is difficult because changes in pollution may be smaller than the influences of meteorology. This paper describes a two-stage modeling approach to estimate trends in concentrations of sulfur dioxide (SO_2) and particulate sulfate (SO_4^{2-}) measured at 33 monitoring sites in the Clean Air Act Status and Trends Monitoring Network (U.S. Environmental Protection Agency, 1998). This approach reduces nontrend variation in the data, including mitigating the effects of meteorology. Monitoring site locations are predominantly rural by design to provide insight into background levels of pollutants where urban influences are minimal. These data are particularly important to study the effect of regional emissions for which long-range transport plays an important role.

2 Data

This analysis is applied to weekly measurements of SO_2 and SO_4^{2-} concentrations ($\mu g/m^3$) at 33 rural long-term monitoring sites in the eastern U.S. that are part of the CASTNet. These data were observed across an eleven-year period from January, 1990 to December, 2000. Continuous measurements of temperature (degrees Celsius), wind speed ($m\,s^{-1}$), and wind direction (degrees clockwise from north) are made at each site. The east-west wind component (u) is calculated as –windspeed× sine(wind direction) and the north-south wind component (v) is calculated as –windspeed× -cosine(wind direction). It was necessary to summarize hourly meteorological data on the same scale as the SO_2 and SO_4^{2-} measurements, i.e., weekly. These meteorological summaries were calculated by averaging all hourly meteorological variables between 10 a.m. and 5 p.m. across the week to characterize conditions during periods of atmospheric mixing. All sites in this analysis were required to have 75 percent of all weeks with concurrent pollutant concentration and meteorological data.

3 Two-stage Model for Regional Trend

The first stage uses a linear additive model to relate the logarithm of weekly SO_2 and SO_4^{2-} concentrations to prevailing meteorological conditions, season and time. The model is of the form

$$\begin{aligned}\log(C_{ijk\ell}) &= \sum_{k=1}^{11} \gamma_{kl} \times \mathbf{1}(\text{year}_k) + \sum_{j=1}^{11} \phi_{j\ell} \times \mathbf{1}(\text{month}_j) + f_{1\ell}(\text{temperature}_{ijk\ell}) \\ &+ f_{2\ell}(\text{humidity}_{ijk\ell}) + g_l(u_{ijk\ell}, v_{ijk\ell}) + \epsilon_{ijk\ell}\ , \qquad (1)\end{aligned}$$

where $C_{ijk\ell}$ refers to the measured pollutant concentration of the i^{th} week within the j^{th} month and k^{th} year at the ℓ^{th} site ($\ell = 1, \ldots, n_s$), $\epsilon_{ijk\ell} \sim N(0, \sigma_l^2)$, $\mathbf{1}$ is an indicator function for the year and the month variables. We represent $f_{1\ell}$ and $f_{2\ell}$ as either simple linear functions or an arbitrary smooth function using splines, which are defined as piecewise polynomial functions with smooth connections between one segment and another. Here, we use cubic B-splines (Green and Silverman, 1994) to model the functional relationships for temperature and humidity. A sum of radial basis functions is used to model the bivariate wind component relationship in (1).

Given our goal of estimating trend in pollutant concentrations adjusted for seasonal and meteorological effects, we use the estimates and variances of the year effect in model (1) to estimate site-specific trends. For each site location, ℓ, the estimated year effect ($\hat{\gamma}_{k\ell}$) is assumed to have a normal distribution with parameters $\gamma_{k\ell}$ and $\sigma^2_{k\ell}$ given the logarithmic transformation used in model (1). Then $\mathrm{E}[\exp(\hat{\gamma}_{k\ell})]=\exp(\gamma_{k\ell}+\sigma^2_{k\ell}/2)$ and site-specific trend ($Z(s_\ell)$) expressed as a percent change between 1990 ($k=1$) and 1999 ($k=10$) may be defined as

$$Z(s_\ell) = 100[\exp\{(\gamma_{10\,\ell} - \gamma_{1\,\ell}) + (\sigma^2_{10\,\ell} - \sigma^2_{1\,\ell})/2\} - 1]. \quad (2)$$

Trends for other annual periods of interest can be estimated by substituting the appropriate estimates of year effects and variances. Although the resulting trend cannot be directly related to emissions *per se*, it seems likely that emission changes would be the dominant effect in the trend. The delta method based on retaining the first term of a Taylor series expansion of a transformation function for trend, $f(\gamma_{1\,\ell}, \gamma_{10\,\ell}, \sigma^2_{1\,\ell}, \sigma^2_{10\,\ell}) = 100[\exp\{(\gamma_{10\,\ell} - \gamma_{1\,\ell}) + (\sigma^2_{10\,\ell} - \sigma^2_{1\,\ell})/2\} - 1]$, is used to approximate the variance of trend at location ℓ and the covariance of trend for different site locations ℓ and ℓ'. In practice, we find the variance terms are so similar that the influence of the difference in these quantities is negligible. Therefore, we do not consider the variance terms in the application of the delta method.

3.1 Second Stage Spatial Prediction of Trends

To make inference about trend at non-monitored locations as well as regional trends, we apply an extension of kriging analysis that allows for the errors of trend estimation in the first stage. We assume there is an underlying and unobserved spatial field $Z(s)$ where $Z(s)$ measures the "true" trend at locations at s. For each monitoring site s_ℓ, we make an observation $\tilde{Z}(s_\ell)$, corresponding to the first stage trend estimate at site ℓ. We assume our regression estimate of trend represents the true trend with error:

$$\tilde{Z}(s_\ell) = Z(s_\ell) + e_\ell \, . \quad (3)$$

Here, $e_\ell \sim N(0, \tilde{\sigma}^2_\ell)$ are interpreted as measurement errors, independent of the random field $Z(s)$. Each monitoring site has its own "nugget effect", $\tilde{\sigma}^2_\ell$, that are assumed known and equal to the first stage estimates of the variances of trends obtained by the delta method. Note that these variances are not the variances of the year effects ($\sigma^2_{k\ell}$) defined in (2). For now, we assume e_ℓ to be independent random variables.

The true underlying process $Z(s)$ is a spatial process consisting of spatial trend and spatially correlated errors. We assume $Z(s)$ is a Gaussian random field $\{Z(s),\ s \in \Re^2\}$, with

$$\mathrm{E}\{Z(s)\} = \sum_{j=1}^{q} \beta_j\, f_j(s); \qquad \mathrm{Cov}\{Z(s), Z(u)\} = \alpha\, K_\theta(||s-u||), \tag{4}$$

where β is a q-vector of unknown regression parameters, $f_1(s), \ldots, f_q(s)$ are known functions of site locations, $\alpha = Var\{Z(s)\}$, $K_\theta(\cdot)$ is an isotropic correlation model parametrized by $\theta \in \Theta$ controlling the range of correlation, and $||s-u||$ denotes either the Euclidean or geodesic distance[1] between site locations s and u. Models (3) and (4) together form a hierarchical model for the "trend data", $\tilde{Z} = (\tilde{Z}(s_1), \ldots, \tilde{Z}(s_n))'$. Given that both parts of the hierarchy are assumed to be normally distributed, we can proceed directly to the likelihood of the model parameters

$$L(\beta, \alpha, \theta; \tilde{Z}) = \left(\frac{1}{2\pi}\right)^{n_s/2} |\Sigma|^{-1/2} \exp\left\{-\frac{1}{2}(\tilde{Z} - X\beta)'\, \Sigma^{-1}(\tilde{Z} - X\beta)\right\} \tag{5}$$

where X is a known $n_s \times q$ matrix (full rank) of known regressors, $X_{ij} = f_j(s_i)$, and Σ is the $n_s \times n_s$ matrix defined by $\Sigma_{ij} = \alpha\, V_{ij}(\theta) + S_{ij}$, $V_{ij}(\theta) = K_\theta(||s_i - s_j||)$ and $S_{ij} = diag(\tilde{\sigma}_1^2, \ldots, \tilde{\sigma}_{n_s}^2)$. An alternative modeling strategy which allows for spatial correlations among the measurement errors could be used. Under this model, the diagonal S would be replaced with a general covariance matrix S =Var(e). We consider three commonly used correlations models: exponential, Gaussian, and spherical. For each model, the parameters β, α (sill), and θ (range) are estimated by maximum likelihood (ML). Further, we consider a geometrically anisotropic form of the correlation model (see Cressie, 1993) with two additional spatial parameters: a dilation parameter, λ, and a rotation parameter, ν.

3.2 Regional Trend Estimation

Simple averaging over monitoring sites within a region may be a poor estimate of regional trend. Kriging methods can be used to produce estimates with minimum mean squared error based on modeling the spatial dependence demonstrated by the data. Since kriging variances do not account for the estimation of unknown spatial covariance parameters, Bayesian techniques are also used to estimate regional trends and standard errors. The, these two approaches are compared to determine the effect

[1]The distance in units of 100 km (D) between two sites with longitude-latitude coordinates in radians $((x_1, y_1), (x_2, y_2))$ is $D = 127.234$ arc $\sin[(\sqrt{T})/2]$ where $T = [cos(y_1)\, cos(x_1) - cos(y_2)\, cos(x_2)]^2 + [cos(y_1)\, sin(x_1) - cos(y_2)\, sin(x_2)]^2 + [sin(y_1) - sin(y_2)]^2$.

of ignoring the uncertainty of the covariance parameters on inference about regional trends.

In the Bayesian analysis using the geometric anisotropic correlation model, we define the posterior distribution of the covariance parameters by use of the likelihood function in (5) with joint prior density

$$\pi(\beta, \alpha, \theta, \lambda) \propto \alpha^{-a_1-1} \exp(-b_1/\alpha)\theta^{-a_2-1} \exp(-b_2/\theta)\lambda^{-a_3-1} \exp(-b_3/\lambda), \quad (6)$$

where now β has an improper prior density assumed constant on $(-\infty, \infty)$, and α, θ and λ all have inverted gamma priors. We also denote the right hand side of equation (6) by $\pi(\alpha, \beta, \lambda)$. In the model we are considering with measurement error, the likelihood tends to a positive constant as any of the covariance parameters tends to 0 or ∞, so an improper prior leads to an improper or highly diffuse posterior distribution. For the isotropic model, we omit the prior density for the geometric anisotropic dilation parameter (λ).

Then $\pi(\beta, \alpha, \theta, \lambda|\tilde{Z}) \propto L(\beta, \alpha, \theta, \lambda)\, \pi(\beta, \alpha, \theta, \lambda)$ and after integrating this posterior with respect to β we obtain

$$\pi(\alpha, \theta, \lambda|\tilde{Z}) \propto \pi(\alpha, \theta, \lambda)\, |\Sigma|^{-\frac{1}{2}} |X'\Sigma^{-1}X|^{-\frac{1}{2}} \exp\left\{-\frac{1}{2}(\tilde{Z} - X\hat{\beta})'\, \Sigma^{-1}(\tilde{Z} - X\hat{\beta})\right\}. \quad (7)$$

For the geometric anisotropic and isotropic models, the posterior distribution is not amenable to analytical treatment, so we use Markov Chain Monte Carlo (MCMC) methods (see Gilks *et. al.*, 1996) to draw samples of the covariance parameters from the respective posterior distribution for inference about Bayesian estimates of regional trends. The overall Bayesian posterior regional means and variances are calculated using iterative expectation formulae described by Holland *et. al.* (2000).

4 Results

4.1 Spatial Covariance

In terms of the Akaike (AIC) and Bayesian Information Criterion (BIC), the two best models for SO_2 appear to have constant mean, general S using geodesic distances and geometrically anisotropic covariance using longitude-latitude coordinates. A comparison of the two best models using the asymptotic result that twice the difference in the negative log-likelihood values is approximately distributed as a chi-square distribution with q degrees of freedom (q is the difference in the number of parameters) shows the isotropic model with geodesic distances would just be accepted ($p-$value=0.052). However, the comparison seems sufficiently close to justify considering the anisotropic as well as the isotropic model in subsequent analyses. For

SO_4^{2-}, a Gaussian correlation model with general S and constant mean function using geodesic distances is nearly identical to the same model with geometric anisotropy in terms of AIC and BIC. Using an asymptotic generalized likelihood-ratio test to compare these models, we observe that the isotropic model with geodesic distances would be just accepted ($p-$value=0.051). Again, it seems reasonable to consider both model in the following analyses.

4.2 Regional Trend Estimates

For the anisotropic covariance, modeling explorations indicated that if the inverted gamma shape parameter (a_j) was too close to 0 or b_j/a_j was too far from the ML estimate of the corresponding parameter (α, θ and λ respectively for $j = 1, 2, 3$), then the mode of the Bayesian posterior distribution for each parameter was some distance from ML estimate of the parameter. To avoid this undesirable effect of the prior distribution, b_j was fixed so that b_j/a_j was equal to the ML estimate of the corresponding parameter. Two classes of priors were considered: (1) "wide priors" with a_1=a_2=a_3=0.1 and (2) "narrow priors" with a_1=a_2=a_3=2.0. In each case, the b_j parameters were selected as described. A similar approach was adopted for the isotropic model where we fixed a=0.1 and a=2. For both pollutants, the choice of the "wide priors" leads to a very diffuse posterior density with a mode that in some cases is well removed from the ML estimate. Given this result, we conclude that the "narrow prior" is superior because it provides results similar to the MLE analysis.

Using the ML estimation and Bayesian techniques, regional trends were obtained for two broad areas in the Midwest (includes Illinois, Indiana and Ohio) and Mid-Atlantic (includes Pennsylvania, Maryland, West Virginia, and Virginia) regions of the U.S. The highest frequency of CASTNet monitoring sites can be found within these regions. Regional trend estimates and standard errors are given in Table 1 for both classes of Bayesian priors, isotropic and anisotropic models, and MLE to show how regional inference might differ with the choice of the underlying covariance model. For both pollutants, the regional estimates of trend for the "narrow prior" class are closer to the ML estimates than those for the "wide prior" class. The Bayesian standard errors are slightly larger than those for the MLE, but it appears that accounting for the uncertainty in estimating the covariance parameters does not significantly increase the regional uncertainties.

In both regions, the estimated regional trends in SO_2 concentrations are consistent with annual SO_2 emissions from affected utility sources participating in EPA's Acid Rain Program established under Title IV of the 1990 CAAA. Estimates of regional trends in SO_4^{2-} were less than those for SO_2 (see Table 1).

5 Conclusions

Site-specific and regional trends in airborne concentrations of SO_2 and SO_4^{2-} are estimated in the presence of variations in atmospheric chemistry and meteorology at 33 rural CASTNet monitoring sites in the eastern U.S. from 1990-1999. These results are useful for evaluating the effectiveness of legislated SO_2 emission control strategies and provide input needed for making informed pollution management decisions over regional-scale landscapes. Significant reductions in SO_2 and SO_4^{2-} emissions under the Clean Air Act Amendments of 1990 have resulted in unprecedented improvements in SO_2 and SO_4^{2-} concentrations.

Disclaimer

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SO_2

Region	Models	MLE	Bayes with "Wide Prior"	Bayes with "Narrow Prior"
Midwest	isotropic	-40.0	-38.4	-39.8
		(3.4)	(3.6)	(3.6)
Midwest	anisotropic	-40.3	-37.9	-40.2
		(3.2)	(3.6)	(3.4)
Mid-Atlantic	isotropic	-33.9	-36.6	-34.8
		(3.6)	(3.9)	(3.8)
Mid-Atlantic	anisotropic	-32.8	-36.8	-34.1
		(3.5)	(3.7)	(3.7)

SO_4^{2-}

Region	Models	MLE	Bayes with "Wide Prior"	Bayes with "Narrow Prior"
Midwest	isotropic	-33.9	-34.6	-34.5
		(4.0)	(4.1)	(4.0)
Midwest	anisotropic	-36.2	-34.9	-35.7
		(3.5)	(3.9)	(3.7)
Mid-Atlantic	isotropic	-31.2	-32.0	-31.6
		(3.6)	(3.7)	(3.7)
Mid-Atlantic	anisotropic	-31.8	-33.2	-32.1
		(3.5)	(3.8)	(3.7)

Table 1: Comparison of regional SO_2 and SO_4^{2-} regional trend estimates (%) and their standard errors (%) (shown in parentheses) for both isotropic and anisotropic model by MLE, Bayes with "wide prior", and Bayes with "narrow prior".