

## Assessing sources of variability in measurement of ambient particulate matter

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### SUMMARY

Particulate matter (PM), a component of ambient air pollution, has been the subject of United States Environmental Protection Agency regulation in part due to many epidemiological studies examining its connection with health. Better understanding the PM measurement process and its dependence on location, time, and other factors is important for both modifying regulations and better understanding its effects on health. In light of this, in this paper, we will explore sources of variability in measuring PM including spatial, temporal and meteorological effects. In addition, we will assess the degree to which there is heterogeneity in the variability of the micro-scale processes, which may suggest important unmeasured processes, and the degree to which there is unexplained heterogeneity in space and time. We use Bayesian hierarchical models and restrict attention to the greater Pittsburgh (USA) area in 1996. The analyses indicated no spatial dependence after accounting for other sources of variability and also indicated heterogeneity in the variability of the micro-scale processes over time and space. Weather and temporal effects were very important and there was substantial heterogeneity in these effects across sites. Copyright © 2001 John Wiley & Sons, Ltd.

**KEY WORDS:** air pollution; hierarchical models; MCMC; measurement error; micro-scale variability; spatial dependence

### 1. INTRODUCTION

Regulation of air pollution has been an important goal of the US Environmental Protection Agency (USEPA) over the last quarter of this century. The impetus for regulation has stemmed, in part, from epidemiological studies that have characterized relations between various forms of ambient air pollution and morbidity and mortality, although not without substantial controversy. Studies in the early and mid-1990s have suggested that particulate matter (PM), a component of ambient air pollution, is related to increased mortality among the elderly and those with cardiovascular and respiratory disease (e.g., Dockery & Pope, 1994; Li & Roth, 1995; Schwartz, 1995). PM ‘is the generic term for a broad class of chemically and physically diverse substances that exist as discrete particles over a wide range of sizes’ (US EPA, OAQPS Staff Paper, 1996). These substances include dust, dirt, soot, smoke and liquid droplets, and are related to emissions from factories, power plants, motor vehicles, construction, fires, and windblown dust. They can also be formed in the atmosphere through chemical reactions under certain weather conditions.

Two sizes of PM have been the object of proposed regulations or standards, those being PM<sub>10</sub>, which refers to all particles with a mean aerodynamic diameter of less than or equal to 10 microns and

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PM<sub>2.5</sub>, all particles less than or equal to 2.5 microns. In this article, we shall focus on PM<sub>10</sub>. Measuring ambient PM<sub>10</sub> accurately is important for regulatory purposes and to assess its relation with health effects. Recently, authors have tried to characterize the spatial component in the analysis of PM data (e.g., Zidek, 1998; Cressie, Kaiser, Daniels, Aldworth, Lee, Lahiri & Cox, 1999).

In this article, we shall explore whether there is heterogeneity in the variability of the micro-scale PM processes across time and space, and the importance of different sources of variability in producing observed PM values. Sources of variability include spatial, temporal, and meteorological effects. In particular, we will consider the degree to which there exists unexplained spatial and temporal heterogeneity. Heterogeneity in the small-scale processes may indicate the presence of important unmeasured processes, including micro-environmental conditions, and possible dependence of the actual observational process on space and/or time.

In Section 2, we shall describe the data available from 1996 in the greater Pittsburgh area, on which our analysis was based, and discuss the results from some preliminary data exploration. Several hierarchical models to characterize sources of variability will be introduced in Section 3. We present the results of applying these models to the Pittsburgh data in Section 4, and Section 5 contains a summary and discussion.

## 2. DATA: PITTSBURGH, 1996

We focus our analysis on the Pittsburgh metropolitan area (USA) in 1996. The Pittsburgh area contains 25 PM monitoring sites, located in a rectangle of about 40 km × 80 km around the city, that report values as 24 h averages (Figure 1). Not all sites report on each day and some sites have multiple measurements ('replicates') on the same day. The number of observations reported ranged from 11 to 33 per day (coming from 10 to 25 sites) for a total of 6448 observations in 1996. A total of 23 weather variables were recorded: the subset selected for our final model will be discussed in Section 2.2. Site-specific observations of weather information were not available and as a result, weather data recorded at the Pittsburgh international airport was used for all sites (Figure 1).

### 2.1. Preliminary Data Exploration

Following Cressie, Kaiser, Daniels, Aldworth, Lee, Lahiri & Cox (1999), who analyzed a portion of the data used here, we employed a logarithmic transformation of PM<sub>10</sub> to stabilize the mean/variance relationship. Figure 2 shows the daily variation of log-transformed PM<sub>10</sub>. The moving average smoother suggests that log (PM<sub>10</sub>) is higher in the summer season, suggesting seasonal effects. It also appears that measurements of PM<sub>10</sub> were higher during the week than on weekends, which could be due to a higher volume of traffic and industrial emissions during the week.

We examined potential spatial dependence of log(PM<sub>10</sub>) with variogram clouds (not shown). The variogram clouds suggested a positive relationship between the magnitude of the variogram and distance between sites, suggesting the presence of spatial dependence. Figure 3 shows heterogeneity across sites in both the level and variability of observed PM values.

We would expect wind and precipitation to have substantial effects on log(PM<sub>10</sub>). This expectation for wind is confirmed in Figure 4. The dotted line in the left plot is a cyclically moving average with a 60° window and the solid line is a least-squares fit of the cosine-transformed wind direction. Precipitation also was important, and precipitation on the previous day explained slightly more variability in PM<sub>10</sub> than precipitation on the current day ( $R^2 = 0.46$  vs. 0.45).

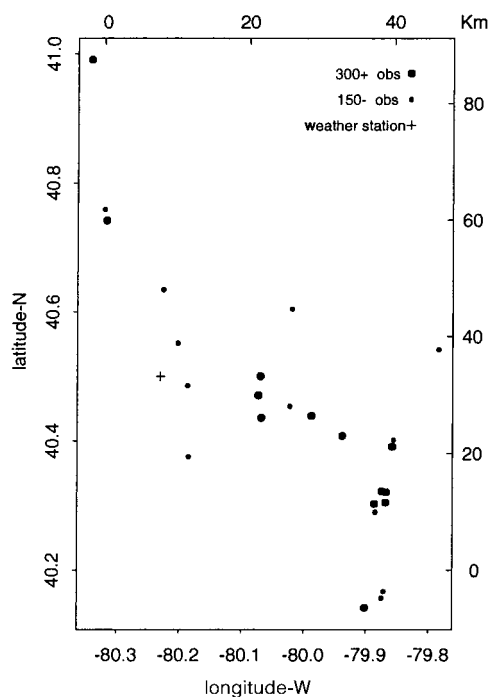


Figure 1. Map of monitoring sites and the weather station (longitude and latitude in degrees)

## 2.2. Time and Weather Variable Selection

The exploratory analysis suggested long or seasonal and short-term temporal effects. To account for noticeable differences between weekday and weekend values (short-term effects), we included an indicator for weekend (WK) in the models. To model the seasonal effects, suggested in Figure 2, we introduced a second order polynomial in time (days).

Preliminary variable selection among the 23 weather variables available in the database was conducted by fitting non-hierarchical linear models. The chosen variables may be divided into three groups: temperature; wind; and precipitation. Based on the preliminary data exploration in Section

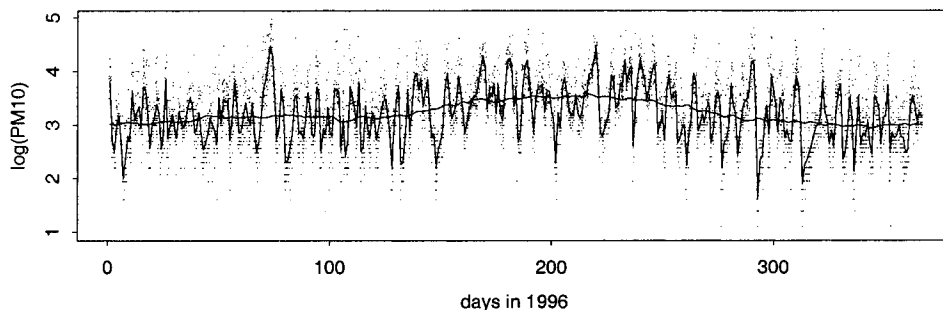


Figure 2. Daily variation of log(PM10). Daily means and a smoothed moving averages are given as solid lines

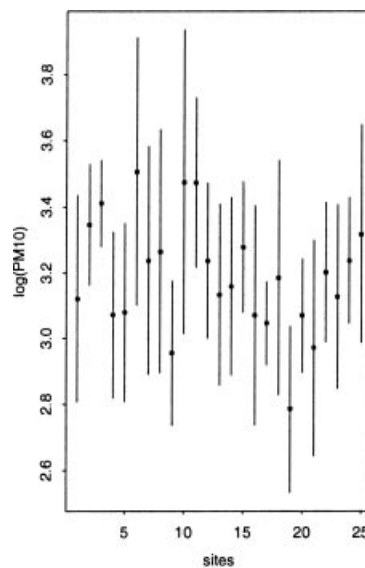


Figure 3. Variation by site. Plotted points are mean levels at site (month) and lines give the middle 95% of the distributions

2.1, we included cosine-transformed wind direction and previous day's precipitation. We also selected five additional weather variables to include in the models examined. From the group of temperature variables, average temperature, dew point temperature (a surrogate for humidity), difference of maximum and minimum temperature, and wet bulb temperature were selected. Another important

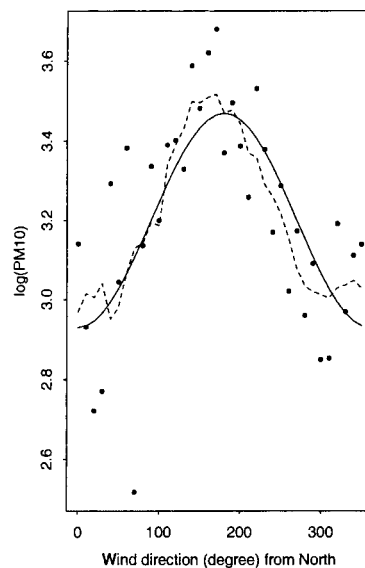


Figure 4. Effects of wind direction. The plot shows the level of  $\log(\text{PM})$  as a function of wind direction. The plotted points are the observed daily means, the dotted line is a cyclically moving average, and the solid line is a least-squares fit using cosine wind direction

wind variable was average wind speed. Thus, we used a total of 10 variables to describe large-scale variation.

### 3. MODELS

Due to the potential importance of time, space, and meteorology in explaining the variability in measured PM<sub>10</sub>, we present several Bayesian hierarchical models to partition the total variability in PM measurements into three components: (1) within sites given time, which includes measurement error and micro-scale atmospheric processes, (2) among sites, which includes spatial heterogeneity in seasonal and weather effects and spatial dependence, and (3) within sites across time, which includes weather and other seasonal patterns and/or temporal dependence. The first and third of these components of variability will be modeled in Stage I of our hierarchical models and the second in Stage II. For all of the models, we consider the time and space components to be separable. The models considered differ in the manner in which the first of these components, the measurement error and micro-scale variability, is treated, and in the inclusion of spatial dependence in the site-specific intercepts in Stage II.

To formulate our models, let  $Z_{ijk}$  be a random variable associated with log(PM<sub>10</sub>) for the  $i$ th location (site),  $j$ th day (of 1996), and  $k$ th replication and define  $\mathbf{Z}_{ij} \equiv (Z_{ij1}, \dots, Z_{ijk_{ij}})$ , where  $k_{ij}$  is the number of values at site  $i$  on day  $j$ .

#### 3.1. Stage I: Within Site Variability

Specify the distribution of the vector  $\mathbf{Z}_{ij}$  as

$$\mathbf{Z}_{ij} \sim N(W_j \alpha_i + X_j \beta_i + \gamma_i, \sigma_{ij}^2 I_{ij}), \quad i = 1, \dots, N, \quad j = 1, \dots, n_i$$

where  $X_j$  is a design matrix of temporal or seasonal effects,  $W_j$  is a design matrix of meteorological or weather effects on day  $j$ , and  $I_{ij}$  is a  $k_{ij}$ -dimensional identity matrix. These design matrices are indexed only by  $j$  since weather data is only available for the whole region for each day. The specific choices for these design matrices was discussed in the previous section. In this model, each location has its own intercept and sets of regression coefficients for temporal and weather effects, namely,  $\gamma_i$ ,  $\alpha_i$ , and  $\beta_i$ :  $i = 1, \dots, n$ . Variability within sites across time is captured in the mean structure of this model stage, namely  $W_j \alpha_i + X_j \beta_i + \gamma_i$ . Within site variability given time is represented by the variance  $\sigma_{ij}^2$ , for site  $i$  on day  $j$ . In the next stage, we will allow variation in the small-scale processes  $\sigma_{ij}^2$  to be constant or to vary over time or space.

#### 3.2. Stage II: Between Site Variability

Let  $\gamma_v \equiv (\gamma_1, \dots, \gamma_n)'$  denote the set of site-specific intercepts, and take

$$\gamma_v \sim N(\gamma \mathbf{1}, \theta^2 (I - C)^{-1}), \quad \alpha_i \sim N(\alpha, D_\alpha), \quad \beta_i \sim N(\beta, D_\beta) \quad (1)$$

The  $C$  matrix was chosen to model potential spatial dependence; for the models with no spatial dependence, we set  $C$  equal to the zero matrix. For models that incorporate spatial dependence, we

construct the prior using a conditional specification (e.g., Besag, 1974; Kaiser & Cressie, 2000). Specifically, we assume isotropic spatial dependence by setting the matrix  $C = C(\eta)$  with zeros for the diagonal terms and off-diagonal elements  $c_{hl} = \eta d_{hl}^{-1}$ , where  $d_{hl}$  is the distance between sites  $h$  and  $l$ .

Temporal effects within a site are modeled through the prior on  $\beta_i$ , which allows for non-stationarity in the temporal covariance structure (Diggle, Liang & Zeger 1994, p. 89). At the same time, the prior induces spatial heterogeneity in the temporal effects. Spatial heterogeneity in the effect of weather variables is modeled through the prior on  $\alpha_i$ . This prior allows the effects of these weather variables to vary across locations due to topography and other micro-environmental conditions.

We consider three models for  $\sigma_{ij}^2$ , the variance of the small-scale processes, which models measurement error and micro-scale atmospheric processes. Our first model takes  $\sigma_{ij}^2 \equiv \sigma^2$ , with no heterogeneity over time or space allowed. Our second model allows the variance of micro-scale processes to change over space by taking  $\sigma_{ij}^2 = \sigma_i^2$  and  $\log(\sigma_i^2) \sim N(\phi_s, \tau_s^2)$ . Our third model allows the variance of micro-scale processes to change over time, by letting  $\sigma_1^2, \sigma_2^2, \dots, \sigma_{12}^2$  denote distinct values indexed by month of the year, and setting  $\sigma_{ij}^2 = \sigma_m^2$  if day  $j$  falls in month  $m : m = 1, \dots, 12$ . While the use of months is arbitrary, this model allows temporal variation in the  $\sigma_{ij}^2$  while assuming that over relatively short time spans of 1 month, variability in small-scale processes is constant. The model is then completed by taking  $\log(\sigma_m^2) \sim N(\phi_t, \tau_t^2)$ . An alternative prior formulation would be to place inverse gamma priors on  $\sigma_m^2$  and  $\sigma_i^2$  (Lin *et al.*, 1997). Micro-scale heterogeneity will not impact the fixed effects ( $\alpha, \beta, \gamma$ ), but will have an effect on prediction of site-specific PM10 level (see Section 4.1 for more details).

Counting these three structures for micro-scale heterogeneity combined with the absence or presence of spatial dependence, we have formulated a total of six models that partition the total variability in PM10 measurements in different ways. The models are all similar in that variability among sites is restricted to be constant over time. To connect the modeling of heterogeneity in micro-scale processes with the original division of variability into three components given at the beginning of this section, parameters relevant to those components of variability are given in Table 1.

To assess the contribution of the fixed weather and temporal effects (specified by the design matrices  $W_j$  and  $X_j$ ), we also fit a reduced model without these effects.

### 3.3. Stage III: Hyperparameters

We utilized non-informative priors for all of the hyperparameters (i.e., parameters of distributions assigned in Stage II of the model). Specifically, on  $\alpha, \beta, \gamma, \phi_t$ , and  $\phi_s$ , normal priors with large variances (approximating a uniform prior) were used, and on the inverses of  $D_\alpha, D_\beta, \theta^2, \tau_s^2$ , and  $\tau_t^2$ , Wishart and Gamma priors were adopted as:

$$D_\alpha^{-1} \sim \text{Wish}(\dim(\alpha), \dim(\alpha), A_\alpha)^{-1}, D_\beta^{-1} \sim \text{Wish}(\dim(\beta), \dim(\beta), A_\beta)^{-1}, 1/\theta^2 \sim \text{Gamma}(2, 2s_c^2)^{-1}, \\ 1/\tau_s^2 \sim \text{Gamma}(2, (2s_s^2))^{-1}, \quad 1/\tau_t^2 \sim \text{Gamma}(2, (2s_t^2))^{-1}$$

Table 1. Model parameters associated with three sources of variability in observations of PM10

Component of variability	Micro-scale heterogeneity		
	No	Spatial	Temporal
Within sites given time	$\sigma^2$	$\phi_s$	$\phi_t$
Among sites	$D_\alpha, D_\beta, D_\gamma$	$D_\alpha, D_\beta, D_\gamma, \tau_s^2$	$D_\alpha, D_\beta, D_\gamma$
Across time	$\alpha, \beta, \gamma$	$\alpha, \beta, \gamma$	$\alpha, \beta, \gamma, \tau_t^2$

For the model with no heterogeneity in the micro-scale processes, we assumed a prior for  $\sigma^2$  to be

$$1/\sigma^2 \sim \text{Gamma}(2, (2s_g^2))^{-1}$$

We choose  $A_\alpha$ ,  $A_\beta$ ,  $s_c^2$ ,  $s_s^2$ ,  $s_t^2$  and  $s_g^2$  as estimates of  $D_\alpha$ ,  $D_\beta$ ,  $\theta^2$ ,  $\tau_s^2$ ,  $\tau_t^2$  and  $\sigma^2$ , respectively, using the data. Thus, these priors are data-dependent.

The prior on the spatial dependence parameter  $\eta$  is assumed to be flat (uniform) in the region where  $(I - C(\eta))^{-1}$  is positive definite. This region can be identified using the eigenvalues of the matrix  $C(\eta)/\eta$  (Cressie, 1993, p. 471). In our case, the region is given as  $(-0.707, 0.408)$ . Estimation is hierarchical Bayes, accomplished using a Gibbs sampling algorithm. Details on the full conditional distributions required for this algorithm are given in Appendix.

## 4. RESULTS

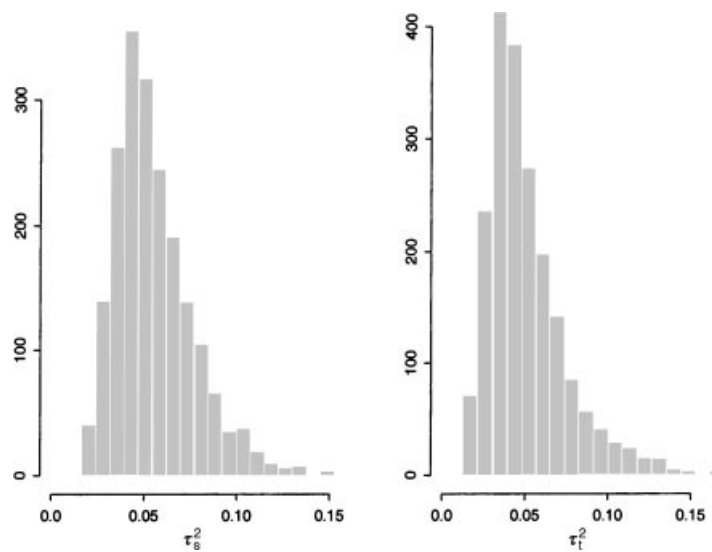
### 4.1. Variability Within Sites Given Time

The presence of heterogeneity in the variance of the micro-scale processes over space or time may be assessed by testing  $\tau_s^2 > 0$  or  $\tau_t^2 > 0$ , respectively, and this may be accomplished using Bayes factors or by examining a prediction error at the observed sites for the different heterogeneity models. For the former, since the Bayes factor depends on the prior for  $\tau_s^2$  ( $\tau_t^2$ ) a prior is needed for  $\tau_s^2$  ( $\tau_t^2$ ), which is relatively 'non-informative' (expressing our ignorance), but allows for the possibility that  $\tau_s^2 = 0$  ( $\tau_t^2 = 0$ ). We use a uniform shrinkage prior (see, e.g., Christiansen & Morris, 1997; Daniels, 1999) of the form:  $\pi(\tau^2) = \delta_c^2 / (\delta_c^2 + \tau^2)^2$ ,  $\tau^2 > 0$ . For details on the choice of the constant  $\delta_c^2$  and computation of the Bayes factor, see Appendix.

We can also evaluate a prediction mean-squared error (PMSE) at the observed sites to compare models since heterogeneity in the variability of the micro-scale processes will impact site-specific predictions and their standard errors. As an example, consider a simplified model with no weather or time effects and only the random intercept,  $\gamma_i$ , with no spatial dependence, but with the variation in the small-scale processes varying over sites,  $\sigma_i$ :  $i = 1, \dots, n$ . The site specific prediction,  $\hat{\gamma}_i$ , conditional on  $\sigma_i^2$  and  $\theta$ , will be  $\hat{\gamma}_i = \bar{Z}_i S_i + (1 - S_i) \hat{\gamma}$ , where  $S_i = (\sigma_i^2 / n_i) / (\sigma_i^2 / n_i + \theta^2)$  and  $\hat{\gamma}$  is an estimate of a common intercept over all sites and the standard error of prediction will be  $\text{SE}(\hat{\gamma}_i) = (n_i / \sigma_i^2 + 1 / \theta^2)^{-1}$ , where  $n_i$  is the number of observations at site  $i$ . Dependence of site-specific predictions on variability of the micro-scale processes,  $\sigma_i^2$ , is apparent. We now define the PMSE to be  $\text{PMSE} = (1 / \sum_i n_i) \sum_i \sum_j (Z_{ij} - Z_{ij}^{\text{pred}})^2$ , where  $Z_{ij}^{\text{pred}} = \gamma_i + W_j \alpha_i + X_j \beta_i$ .  $Z_{ij}^{\text{pred}}$ , and consequently, PMSE, can be computed at each iteration of the Gibbs sampler.

The posterior distributions of  $\tau_s^2$  and  $\tau_t^2$  appear in Figure 5. Both the spatial and temporal models indicated heterogeneity in the micro-scale variance, with Bayes factors larger than 100 in favor of the models with heterogeneity. The Bayes factor to compare the temporal and spatial models gave a Bayes factor of 2.6, minimal support for spatial heterogeneity. The PMSE at each site was the same for all three models (last row of Table 2). Although in this case the PMSE suggests little difference in overall predictions for the three models, the Bayes factor results strongly supports the presence of heterogeneity in the variability of the micro-scale processes over space and time.

The posterior means of  $e^{\phi_s}$  and  $e^{\phi_t}$  in spatial heterogeneity model and temporal heterogeneity model appear in Table 2. The posterior means reflect about a 15% multiplicative error on the original PM10

Figure 5. Posterior distributions of  $\tau_s^2$  and  $\tau_t^2$ 

scale. We also note that the models discussed in Section 2 have significantly smaller values of  $\epsilon^\phi$ ,  $\sigma^2$ , and PMSE than the models without temporal and meteorological effects (cf. Table 3). This reflects the importance of the inclusion of time and weather effects, which will be discussed more in the next two sections. We also note that the measure of spatial heterogeneity,  $\tau_s$  tends to be larger than the corresponding values for temporal heterogeneity,  $\tau_t$  and that this heterogeneity corresponds to multiplicative errors at individual sites ranging from about 5% to 40% on the original PM scale.

Table 2. Posterior means and 95% credible intervals for the spatial and heterogeneity parameters and PMSE

Parameter	No Hetero,		Spatial Hetero,		Temporal Hetero,	
	mean	95% CI	mean	95% CI	mean	95% CI
$\eta$	-0.135	(-0.655, 0.358)	-0.137	(-0.649, 0.354)	-0.154	(-0.664, 0.359)
$\epsilon^\phi (\sigma^2)$	0.145	(0.140, 0.150)	0.132	(0.117, 0.148)	0.142	(0.122, 0.163)
$\tau (\tau_s, \tau_t)$			0.249	(0.175, 0.353)	0.245	(0.159, 0.386)
PMSE	0.141	(0.140, 0.142)	0.141	(0.140, 0.142)	0.141	(0.140, 0.142)

Table 3. Posterior means and 95% credible intervals for spatial and heterogeneity parameters and the PMSE in the reduced model, without time and weather effects

Parameter	No Hetero,		Spatial Hetero,		Temporal Hetero,	
	mean	95% CI	mean	95% CI	mean	95% CI
$\eta$	0.223	(-0.263, 0.401)	0.226	(-0.246, 0.401)	0.225	(-0.275, 0.401)
$\epsilon^\phi (\sigma^2)$	0.316	(0.306, 0.328)	0.278	(0.245, 0.310)	0.310	(0.273, 0.351)
$\tau (\tau_s, \tau_t)$			0.252	(0.172, 0.359)	0.209	(0.135, 0.328)
PMSE	0.312	(0.312, 0.313)	0.312	(0.311, 0.313)	0.312	(0.312, 0.313)



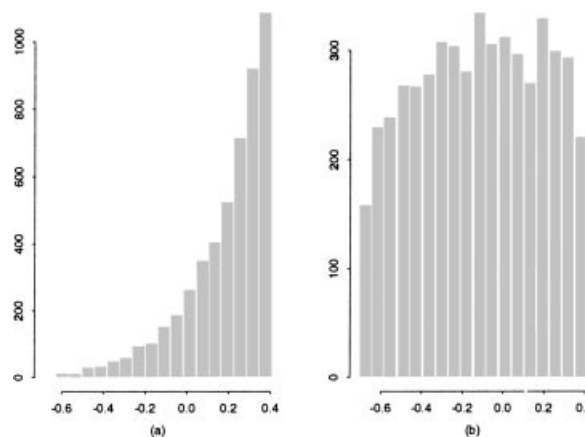


Figure 6. Posterior distributions of  $\eta$ . The left plot (a) shows the posterior in the reduced model, without temporal and meteorological effects, and the right plot (b) shows the posterior distribution in the model with temporal and meteorological effects

#### 4.2. Variability Among Sites

To examine variability among sites, we assess the heterogeneity in the weather and temporal effects, and spatial dependence. To assess the heterogeneity, we examined the variability of the site-specific coefficient,  $\alpha_i$  and  $\beta_i$  (not shown in tables). These coefficients, along with the matrices  $D_\alpha$  and  $D_\beta$ , indicated substantial heterogeneity across sites. For example, the effect of weekend (as opposed to weekday) on levels of PM<sub>10</sub> ranged from about a 13% decrease to a 25% decrease in the levels across sites. For previous day's precipitation, the effects ranged from a 25% to 37% decrease.

To assess spatial dependence, we examined the posterior distribution of  $\eta$ . For all models, 95% credible intervals for  $\eta$  cover zero, implying non-significant spatial dependence (Table 2 and Figure 6). The models with weather and time effects offer little suggestion of isotropic spatial dependence; the posterior distribution of  $\eta$  is very similar to the uniformly distributed prior. However, models without temporal and meteorological effects (Table 3) show a much more peaked posterior distribution for  $\eta$ . Any spatial dependence that is present has likely been explained by the heterogeneity across space modeled in the mean effects ( $\alpha_i$  and  $\beta_i$ ). Future work might assume  $\alpha_i$  and  $\beta_i$  are not exchangeable across sites, but follow a distribution having a spatial dependence structure.

#### 4.3. Variability Within Sites Across Time

Table 4 presents posterior means and 95% credible intervals for the intercept ( $\gamma$ ) and the fixed weather and temporal effects ( $\alpha$  and  $\beta$ ). The estimates were very similar across the models; thus, Table 4 only presents the results for the spatial heterogeneity model. In terms of the weather effects, resultant wind direction was significant and indicated higher levels of PM<sub>10</sub> when the wind blew from the south (cf. Figure 4). This may reflect the effect of wind blowing from an industrial region. A binary indicator of precipitation on the previous day was also significant and indicated lower levels of PM<sub>10</sub> when there was rain on the previous day. PM<sub>10</sub> levels were higher during warmer weather, lower when the dew point temperature (a surrogate for humidity) was high, and lower during periods of high winds. The seasonal effects indicated a peak in PM<sub>10</sub> levels during the summer months and a reduction in PM<sub>10</sub> levels during the weekend. The importance of these effects is also clear from the PMSE which

Table 4. Posterior means and 95% credible intervals for the overall meteorological and time effects

Variable	Mean	95% interval
Intercept	2.70	(2.62, 2.77)
Average temperature	0.030	(0.026, 0.034)
Dew point temperature	-0.013	(-0.017, -0.009)
Daily max. difference of temperature	0.004	(0.001, 0.007)
Cos(wind direction)	-0.160	(-0.213, -0.104)
Wind speed	-0.064	(-0.071, -0.057)
Precipitation	-0.282	(-0.348, -0.224)
Weekend	-0.139	(-0.177, -0.097)
Linear spline	-2.96	(-3.64, -2.30)
Quadratic spline	2.97	(2.50, 3.43)

decreases by over 50% when the weather and temporal effects are included in the model (cf. Tables 2 and 3).

## 5. DISCUSSION

We have proposed several hierarchical models to account for the different sources of variability in monitoring PM10. Our analysis indicates there was no isotropic spatial dependence after accounting for other sources of variability. But, there was an indication of heterogeneity in the variability of the small-scale processes over time and space, and heterogeneity in the covariate effects (and mean values) across sites, indicating the presence of significant unmeasured processes. The heterogeneity in the weather effects across sites might be related to our use of regional weather variables which have the same values for all sites. Though typically not available, the inclusion of site-specific weather covariates would be expected to help to explain this heterogeneity. Heterogeneity will be important for predictions at observed sites and for removing the measurement error from the true PM10 process in general; our models are well suited to give “error-free” predictions at the observed sites and to fill in PM10 values for unmeasured days at those sites. Components of temperature wind, and precipitation, were shown to be important meteorological predictors of PM10 and there appears to be substantial heterogeneity in these effects across sites as well.

The goals of this study were to quantify the different sources of variability in observations of PM10 and to examine their relative importance. However, models of the type developed here could also be used in conjunction with models for health effects. For example, fitted values from this model could be used as a covariates in health effects models which examine the relationship between PM10 and mortality; the standard deviation of the fitted value would provide a measure of uncertainty for the PM10 values. As discussed in Section 4, predictions and their uncertainty will be effected by the heterogeneity in the variability of the small-scale processes. By modelling the variability of the small-scale processes (including measurement error) as we have done, the prediction of PM10 will include an adjustment for the ‘measurement error’; this adjustment should result in increases in the accuracy of estimated effects of PM10 on health (e.g., Rosner, Spiegelman & Willet, 1993).

## APPENDIX

This appendix has two sections. In the first part, we describe the forms of the full conditional distributions used in the Gibbs sampling algorithm and approaches to generate from these full

conditionals. In the second part, we discuss the choice of priors for  $\tau_s^2$  and  $\tau_t^2$  and the relevant computations for the Bayes factors.

### A.1. Full Conditional Distributions

The full conditional distributions for  $\alpha_i$ ,  $\beta_i$ ,  $\gamma_v$ ,  $\alpha$ ,  $\beta$ , and  $\gamma$  are all normally distributed. The full conditionals for the covariance matrices  $D_\alpha$  and  $D_\beta$  are distributed as inverse-Wishart distributions. For the spatial (temporal) heterogeneity model, the full conditional distributions for  $\phi_s$  ( $\phi_t$ ) are normal and for  $\tau_s^2$  ( $\tau_t^2$ ) are inverse-Gamma distributions. For the model without heterogeneity,  $\sigma^2$  follows an inverse-Gamma distribution. In the models with and without spatial dependence in the intercepts,  $\theta^2$  follows an inverse-Gamma distribution.

We use Metropolis–Hastings algorithms to sample from the full conditional distributions of  $\eta$  (in the spatial dependence models), and from  $\sigma_i^2$  ( $\sigma_m^2$ ) in the spatial and temporal heterogeneity models, respectively.

### A.2. Priors and Computations for Bayes Factors

This section provides details on the choice of the prior and the computations for the Bayes factors. We follow Daniels (1999) and choose  $\delta_c^2$  to be the harmonic mean of the sampling variances of  $\log(\hat{\sigma}_p^2)$ :  $p = 1, \dots, n(12)$  for the spatial and temporal heterogeneity models, respectively. These are obtained by fitting non-hierarchical models by site and month, respectively, and setting them equal to the residual variances. Recognizing that asymptotically  $(N_p - 1)\hat{\sigma}_p^2 \sim \sigma_p^2 \chi_{N_p-1}^2$ , the variance of  $\log(\hat{\sigma}_p^2)$  will be  $2/(N_p - 1)$ , where  $N_p$  is the number of observations at the  $p$ th site (in the  $p$ th month). So, we set  $\delta_c^2 = 2/(N - P)$ , where  $P = n(12)$  and  $N = \sum_p N_p$ .

Following the approach in Daniels & Hughes (1997), we can compute the Bayes factor using the Savage–Dickey density ratio (Verdinelli & Wasserman, 1995). In this case, the Bayes factor will be equal to  $p(\tau^2 = 0 | Z)/p(\tau^2 = 0)$ , the ratio of the posterior density to the prior density at  $\tau^2 = 0$ . The numerator can be computed via kernel-density methods.

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