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The Ability of Wet Deposition Networks to Detect Temporal Trends

Gary W. Oehlert*

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May 1994

Abstract

We use the spatial-temporal model developed in Oehlert (1993) to estimate the detectability of trends in wet-deposition sulfate. Precipitation volume adjustments of sulfate concentration dramatically improve the detectability and quantifiability of trends. Anticipated decreases in sulfate of about 30% in the Eastern U.S. by 2005 predicted by models should be detectable much earlier, say 1997, but accurate quantification of the true decrease will require several additional years of monitoring. It is possible to delete a few stations from the East without materially affecting the detectability or quantifiability of trends.

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Careful siting of new stations can provide substantial improvement to regional trend estimation.

1 Introduction

Oehlert (1993) described a spatial/temporal model for wet deposition data in eastern North America. This model includes spatial and temporal correlation among the monitoring stations and provides a way to estimate regional values from a scattering of stations by using a discrete smoothing prior. In this paper, we use the model of Oehlert to estimate the detectability of regional trends in sulfate wet deposition for current and modified networks, based on hypothesized future trends and variances and other model parameters observed to date. In particular, we investigate the detectability of trends in sulfate concentration for seven regions of the Eastern US: Northeast less Appalachians (NE), Mid Atlantic (MA), Appalachians (AP), Midwest (MW), Plains (PL), South (SO), and Mississippi Delta (MD). These regions of the US are shown in Figure 1, as well as four additional regions for the remainder of the continental US: Inter-mountain/Desert (ID), Southwest California (SC), Pacific Northwest (PN), and Rocky Mountains (RM).

The Regional Acid Deposition Model (RADM) (Chang et al, 1990) predicts sulfate wet deposition over eastern North America on the basis of emissions and meteorology. Sulfur emissions are expected to decline over the next 15 years as the result of the 1990 Clean Air Act Amendments. Applying RADM to the anticipated emissions reductions results in approximately 35% reductions in sulfate wet deposition in the Northeast, and less elsewhere. The anticipated changes are not linear over time, but are greatest near the years 1995 and 2002 when the Clean Air Act has reduction goals. While not homogeneous over time or space, we will treat the future sulfate changes as the product of specific regional mean reductions (given in Table 1) and the temporal shape shown in Figure 2 (R. Dennis, personal communication).

We wish to know how well such trends can be estimated using data from the existing NADP/NTN monitoring network. For example, we might ask with what probability could we detect a decrease by the year 1995 (detectability), or with what probability will our estimated change for a region be within 20% of the true value (quantifiability)? These criteria depend on the size of the true regional trend and the variance of our estimate of the regional trend. The main problem is thus to determine the variance of regional trend estimates for response variables of interest (such as sulfate concentration and precipitation-adjusted sulfate concentration).

The structure of this paper is as follows. The next section introduces the data sets we use to determine the variance parameters in the model and discusses the precipitation adjustment. Section 3 gives our results for dectectability and quantifiability for current networks, and Section 4 considers the addition and deletion of stations to the current network. An Appendix briefly reviews the spatial/temporal model of Oehlert(1993) and discusses the computation of detectability and quantifiability.

2 Variance parameters and the data

Detectability and quantifiability depend on the variance of regional trend estimates, which in turn depend on the change scenario $\mathbf{t}$, the station locations, and several variance parameters in the model (the spatial and continental variances S and D and the prior variance $1/\lambda_\beta$) as described in the Appendix. This section discusses the choice of these parameters. As discussed below, all data are analyzed on the log scale, so that coefficients of change are approximately proportional changes.

The temporal change scenario $\mathbf{t}$ is shown in Figure 2. North America is tiled with small rectangles, one degree of latitude by 1.5 degrees of longitude. We assume that every station in rectangle j has the same true change β_j, and the procedure described in the Appendix shows how to estimate the β_j values. Regional estimates are the unweighted averages of the rectangle estimates. The prior variance $1/\lambda_\beta$ (for neighboring rectangle differences) was set to 0.01, with the interpretation that the difference in coefficients between two neighboring rectangles should be zero, plus or minus about 0.10. Given that the anticipated coefficients range fairly smoothly from about 0.15 to 0.35 across eastern North America, this represents a mild smoothing.

We model the spatial covariance S (between stations) using an exponential model. Let S be σ_N^2 on the diagonal and $\sigma_N^2 b \exp(-c\ d)$ for two stations at a distance d. This model of S corresponds to a stationary spatial covariance (with nugget) that is frequently used in Kriging. The continental component variance D is modeled as σ_L^2 times the ARMA(1,1) correlation structure described in the Appendix. Fitting of σ_N, b, c, and σ_L^2 from data is discussed in Oehlert (1993). We note that σ_L^2 is poorly estimated.

Our data come from the ADS data base (Watson and Olsen 1984). We obtained monthly sulfate concentration data for the years 1978 through 1987 and stations in the NADP/NTN, APIOS-C, and CAPMON networks, and form volume weighted annual means. Values are imputed for missing months (see Oehlert 1993 for details), and a station annual mean is missing if precipitation volume measurements are available for less than 90% of the year or concentration values are available for less than 60% of the collected precipitation. For variance parameter estimation, we use all stations from these networks that have no missing years in the period 1982-1986 for the variable of interest. This is the 82-86 station set, denoted in Figure 3 by a $\times$. A second station set comprising all stations in these three networks in operation at the end of 1987 will be used for computing variances of regional averages; these stations are denoted by a $+$ in Figure 3. We assume that the complete station set has the same variance structure as the complete-data station set and use the log of annual concentration, as this scale helps stabilize interannual variability across stations.

Sulfate concentrations tend to be smaller for larger precipitation volumes, due in part to a dilution effect. We will also consider precipitation adjusted concentrations, where the adjustment is as follows. For each station, fit a linear model including a separate mean for each of the 12 calendar months and a common (across months) linear effect for precipitation volume. Let $\hat{\gamma}$ be the estimated coefficient for precipitation volume. For a given monthly concentration, say a value in calendar month i with precipitation volume p_{ij}, form the precipitation adjusted concentration by subtracting $\hat{\gamma}(p_{ij} - \bar{p}_i)$ from the monthly concentration, where $\bar{p}_i$ is the mean precipitation in calendar month i. This adjusts each observation in a given calendar month to a standard precipitation volume. Form adjusted annual concentrations by taking volume weighted means of the adjusted monthly concentrations.

3 Sulfate trend detectability and quantifiability

For unadjusted log sulfate concentration, we have the following parameter estimates: $\sigma_L^2 = 0.00228$, $\sigma_N^2 = 0.0196$, $b = 0.139$, and $c = 0.00194$ (1/km). Figure 4 shows the standard error of the regional estimates of total reduction in log sulfate concentration in 2010 based on our assumed decrease scenario for data years 1995 through 2010. Large regions with many stations (eg., the Plains, Midwest, or South) have smaller standard errors than small regions with few stations (eg., Mississippi Delta or mid-Atlantic). Standard errors decrease nicely over time, with larger decreases in standard error occurring in years where we expect larger changes sulfate concentration. In 2000, a typical standard error for the estimated decrease is about 6 or 7 percent of total starting concentration. This decreases to about 4 percent in 2010.

The detectability of the RADM predicted decrease is excellent, with the five easternmost regions having power essentially one by 1998. The Plains reaches a power of nearly one in 2002, and the Mississippi Delta only reaches power 0.8 by 2003. These regions have lower powers due to smaller anticipated changes, and, in the case of the Mississippi Delta, larger standard errors. Figure 5 shows quantifiability of the estimated 2010 decrease based on data from 1995 through 2010 for the seven Eastern regions, that is, the probability that the estimated regional average decrease is within $0.2\bar{\beta}$ of the true regional average decrease $\bar{\beta}$. Quantifiability is much smaller than power, with the five Easternmost regions not achieving 0.8 quantifiability until 2002.

The contribution of the continental component σ_L^2 to the variance is not well estimated. Thus it is prudent to determine how sensitive our results are to inaccuracies in that variance component. To explore this sensitivity, we computed the regional variances, powers, and quantifiability for the data year 2000 when the continental component is 1) set to zero, 2) used as estimated, and 3) set to twice the estimated value. In the seven Eastern regions, the estimated continental component is responsible for approximately two thirds of the regional standard deviation. Detectability in data year 2000 is not strongly influenced by this modified continental component except for the Plains and the Mississippi Delta, which lose about 15% power when the continental component is doubled. Quantifiability is much more strongly affected by changes in the continental contribution. Removing or doubling the continental component can change regional quantifiability from 5 to 35% depending on the region; see Figure 6.

We now reanalyze using variance estimates based on log precipitation-adjusted sulfate concentrations. The estimated variance parameters are $\sigma_L^2 = 0.00044$, $\sigma_N^2 = 0.0156$, $b = 0.130$, and $c = 0.00224$ (1/km). Thus the short term variance has decreased slightly relative to unadjusted sulfate, but most importantly, the continental component is smaller by a factor of 5 for the adjusted sulfate. Figures 7 and 8 display standard errors and quantifiability using precipitation adjusted variance estimates. Regional variances are about a factor of 2 to 3 smaller using precipitation adjusted data than they were with unadjusted data. All Eastern regions except Mississippi Delta have power essentially one by 1997, and the Mississippi Delta region has power greater than 0.9 by 2001. Quantifiability is also dramatically greater for the precipitation adjusted data, with the five Easternmost stations having quantifiability greater than 0.8 by 1997. These improvements are primarily the result of the smaller estimated continental component.

4 Adding and deleting stations

Monitoring networks are not static; stations are added and stations drop out over time. With tighter budgets, it is sensible to ask questions such as "Given that we have funding to add 10 stations to the network, where should we put them for maximal improvement of the network?" or "Given that we must delete 10 stations, which 10 stations should we delete to have minimal effect on the capabilities of the network?" Of course, the answers to these questions depend on many factors, including which chemical is being studied, whether we are interested in mean or trend, and how we choose to measure the quality of information produced by a network.

We have decided to concentrate on the estimation of change through 2010 in log precipitation-adjusted sulfate concentration for the five Easternmost regions using the RADM change scenario shown in Figure 2. (Similar results are obtained for unadjusted sulfate.) We will use two different criteria for evaluating the quality of a set of stations. The first criterion is the sum of the regional variances for the five Easternmost regions; the second is the sum of the 40 largest variances corresponding to rectangles in the five Easternmost regions. The first criterion concentrates on minimizing the average regional variance, while the second tries to keep the largest local variances from getting too big. The choice of 40 rectangle variances was somewhat arbitrary; they are about one fifth of the rectangles in the five regions.

Figure 9 shows the locations of the 40 largest rectangle variances when using all stations in operation at the end of 1987; Table 2 gives the regional variances and the two criteria. We see that the large variances tend to be along the coast where stations are few and there are no stations "on the other side" to be included in smoothing.

Suppose now that we must delete 10 of the stations from the East, but that Appalachian stations are precious and none will be deleted. Which stations could be deleted with minimal increase in our criteria? To minimize the computations, we have chosen these stations sequentially. That is, we look for the station which least increases the criterion. Then, given that station is deleted, we find which station to delete in addition to the first, and repeat this till we have chosen 10 stations for deletion. (This is the so called greedy algorithm.) In fact, at each stage we have carried forward 10 sets of stations rather than a single set of stations, so that we are more likely to find a good set.

Figures 10 and 11 show the locations of 10 stations for deletion using the greedy algorithm when selected according to the regional variance and rectangle variance criteria respectively. Table 2 gives the criteria values. Both Figures show that most of the deleted stations would come from the Midwest/Great Lakes region, where stations are relatively dense and surrounded by stations in other regions or Canada. When the regional criterion is used, some of the deleted stations are near rectangles with high variance; this occurs to a lesser extent when the criterion is rectangle variance. With both sets of stations and criteria, the deletion of stations has resulted in little increase in the criteria for adjusted sulfate. We note that there are several other sets of stations that give nearly identical results.

Conversely, suppose that we can add 10 stations to the network in any rectangle of the five Eastern regions; where should we add these stations to most reduce our criteria? We have tested the centers of all rectangles as potential additional sites. In reality, the sites would need to be located somewhere within the rectangle (in particular, the coastal sites would need to be on land, not in the ocean!), but we have used the centers as an approximation. Furthermore, we have estimated the improvement in variance as if the stations had data during the entire period 1985-2010, not beginning part way through. This approximation is adequate for showing the general outlines of the results. As before, we add stations sequentially.

Figures 12 and 13 show the locations of the 10 stations to be added when selected according to the regional and rectangle variance criteria using the greedy algorithm; Table 2 again gives the criteria values. Basically all station lie along the East coast. When selecting for regional variances, all new stations are in the Northeast and Middle Atlantic regions; when selecting for rectangle variances, the stations are spread along the coast from Florida to Maine. Looking at the criteria, we see that we can reduce the variance sums by 10 to 15%. However, selecting for the different criteria has resulted in rather different station configurations, and the configurations perform differently for the different criteria.

5 Discussion

The key to any power analysis of this sort is a proper estimate of the variance. The most unusual aspect of our variance model is the inclusion of a continental scale variance component. While the appropriate method for modeling this type of variation is subject to discussion, the existence of long term phenomena in meteorological series, specifically precipitation series, seems accepted, and there is some evidence to suggest that this long term phenomenon is also broad in spatial scale. Thus, wet deposition and, to a lesser extent, concentration series should inherit at least some long term, broad spatial scale variation through their dependence on precipitation volume. Unfortunately, five years of data provide us with a very poor estimate of this continental scale component of variation. For example, the σ_L^2 we estimated for sulfate concentration seems high relative to the overall variability of the series. More years of baseline data will help reduce the variation in our estimates, but for now we must explore the sensitivity of our results to σ_L.

Using either adjusted or unadjusted sulfate, we have an excellent chance of detecting the reductions in sulfate concentration anticipated for the Eastern US as early as the year 1997. This remains true even if we have underestimated the variance of the continental scale component by a factor of two. However, precise quantification of the amount of decrease is more difficult than detection, and will require several more years of monitoring.

We have made precipitation volume adjustments to concentration values using linear regression on monthly data. Better adjustments, including more complicated dilution or meteorological models, should be possible using weekly or event data. For example, Steyer and Stein (1992) investigated meteorological adjustments to wet deposition data used in trend analysis. However, obvious alternatives to our simple approach (such as using log transformed concentrations and precipitation volumes) did not appreciably improve our precipitation adjustment when using monthly data.

The redesign of a network requires some criterion for optimization. We have used two criteria: the local and regional variances of trend estimates. Many other criteria are possible, and these other criteria may lead to different recommendations. For example, Guttorp et al. (1992) argue that we should minimize the entropy of the unobserved data given the observed data. For the network augmentation problem, they found that this criterion (for certain prior distributions) translates to maximizing the determinant of the covariance of the augmented stations given the existing stations. This is not equivalent to our 40 largest rectangle variance criterion, but both will tend to add stations where the local variance given the existing stations is large.

The continental scale variance σ_L^2 does not affect the selection of stations for deletion or inclusion, as σ_L^2 enters only through a constant in the covariance matrix. The prior smoothness precision λ_β could affect the choice of stations, but preliminary investigations show that the dependence on λ_β is fairly weak.

We should note that the recommendations for station deletion and addition assume that the remainder of the stations remain and that the optimization criteria capture our interest in the phenomenon. For example, we would be less likely to remove stations around the Great Lakes if stations in Canada were also being removed. Also, we have been working with log transformed data, and this will tend to emphasize the variance at low concentration stations relative to high concentration stations. This is undesirable if trend at low concentration sites is not of interest.

The station addition results point to questions of station siting that go beyond the variance criteria to the definition of what is being measured. All the sites for addition were near the coast, yet most network siting criteria specifically avoid coastal areas. This avoidance means that coastal estimates could contain a substantial bias, since coastal precipitation is not actually sampled. The addition of stations along the coast would reduce that bias (as well as reduce the estimation variance), but alter what the network measures to include coastal precipitation. This would be important, if, for example, we wished to estimate acid deposition in estuarine ecosystems. If coastal sites are not eligible for measurement, then the coastal regions should not be included in the selection computation and other sites, presumably near coastal, would be selected. Similar problems exist for high elevations.

6 Appendix

Here we present a sketch of the statistical model; see Oehlert (1993) for details. Tile Eastern North America with small rectangles, one degree of latitude by 1.5 degree of longitude. We assume that all stations within a rectangle have the same expected trend, but that trend may vary from rectangle to rectangle. We take as regional trends the unweighted mean of the estimated rectangle values for each of the rectangles in the regions of interest. The variance of these regional average estimates is determined from the covariance matrix of the rectangle estimates.

There are s monitoring stations each with y years of data. Let Y_i be a vector containing the series of annual values at station i. The values could be concentrations, precipitation adjusted concentrations, depositions, etc. Let $j(i)$ indicate the rectangle in which station i occurs. We assume that each series Y_i has the structure

$$Y_i = \alpha_{j(i)}\mathbf{1} + \mathbf{t}\beta_{j(i)} + L + N_i + \delta_i\mathbf{1}, \tag{1}$$

where $\alpha_{j(i)}$ is the mean value in rectangle $j(i)$, $\mathbf{t}$ is the trend shape in Figure 2 (centered to have mean zero), $\beta_{j(i)}$ is the change for rectangle $j(i)$, L is a long-term noise series common to all stations, N_i is a short-term station specific noise series, and δ_i is a station specific random effect accounting biasing effects such as elevation or proximity to point sources of sulfur. Let N denote the vector of all the station specific noise terms, let δ be the vector of station biases, and let $\alpha_J = (\alpha_{j(1)}, \cdots, \alpha_{j(s)})'$ be the vector of rectangle means. The analogous vector for slopes is β_J.

We assume that all station specific noise terms have the same temporal correlation structure, and we assume that the cross covariance factors into a spatial term and a temporal term, so that the covariance matrix of N is

$$Cov(N) = S \otimes C, \tag{2}$$

where S is the spatial covariance matrix and C is the common temporal correlation matrix. Because of the structure of the short term noise series, C is essentially tridiagonal, with sub- and super-diagonal elements nearly zero, say about 0.01. The covariance matrix of L is denoted by D. The station random effects δ_i are uncorrelated with variance σ_δ^2, and we assume that N, L, and δ are uncorrelated. Thus the covariance of Y is

$$\mathbf{1}_{s\times s} \otimes D + S \otimes C + \sigma_\delta^2 I_{s\times s} \otimes \mathbf{1}_{y\times y}. \tag{3}$$

With this notation, observed station slopes (calculated via ordinary least squares) can be expressed

$$b = \beta_J + \mathbf{1}_{s\times 1} \times (\mathbf{t}'\mathbf{t})^{-1}\mathbf{t}'L + I_{s\times s} \times (\mathbf{t}'\mathbf{t})^{-1}\mathbf{t}'N, \tag{4}$$

with covariance matrix

$$Cov(b) = \mathbf{1}_{s\times s} \times (\mathbf{t}'\mathbf{t})^{-1}\mathbf{t}'\ D\mathbf{t}(\mathbf{t}'\mathbf{t})^{-1} + S \otimes (\mathbf{t}'\mathbf{t})^{-1}\mathbf{t}'\ C\mathbf{t}(\mathbf{t}'\mathbf{t})^{-1}. \tag{5}$$

We now estimate the β_j for all rectangles j. The estimates b_i serve as the observations in a spatial linear model. An observed trend b_i in the $j(i)$th rectangle from station i has expected value $\beta_{j(i)}$. We may express this in matrix form as

$$E(b) = W\beta, \tag{6}$$

where the matrix W is all zero except for a 1 in each row indicating rectangle.

In addition to the observed trends, we also use a discrete smoothness prior for β, because we believe that β varies slowly in space. We make this belief explicit by using a partially improper normal prior on β with mean zero and inverse covariance matrix $\lambda_\beta A'A$. The matrix A has a row for every pair of adjacent rectangles, and a column for every rectangle. A is all zeros, except that each row has entries of 1 and -1 for the associated rectangle pair coefficients. The smaller the prior variance for these differences $(1/\lambda_\beta)$, the smoother the resulting estimates will be.

If Σ_b is the covariance matrix of b, then the estimate $\hat{\beta}$ can be expressed

$$\hat{\beta} = (W'\Sigma_b^{-1}W + \lambda_\beta A'A)^{-1}W'\Sigma_b^{-1}b \tag{7}$$

with posterior variance matrix

$$Cov(\hat{\beta}) = (W'\Sigma_b^{-1}W + \lambda_\beta A'A)^{-1}. \tag{8}$$

We use an ARMA(1,1) type correlation structure for D: $\rho_k = \rho_1\phi^{k-1}$ for lags $k \geq 1$. Boes and Salas (1978) show that several models proposed for long term dependence in precipitation records have this ARMA(1,1) covariance structure. Oehlert (1993) used the parameters $\rho_1 = 0.3$ and $\phi = 0.95$, based on analysis of historical precipitation records, and these values will also be used in this study. The variance of this process, σ_L^2 is estimated using covariance between stations at great distances, as discussed in Oehlert (1993).

Regional trend detectability and quantifiability depend on the regional trend and the variance of the estimated trend. Detectability is power. We compute detectability based on the null hypothesis of no trend, using a one-sided test at the 5% level. The power is then computed as $\Phi((\bar{\beta} - 1.645\sigma_r)/\sigma_r)$, where $\bar{\beta}$ is the magnitude of the hypothesized regional trend, σ_r is the standard deviation of the regional trend estimate, and Φ is the standard cumulative normal. To compute quantifiability, we assume that the regional trend estimate is unbiased with mean equal to the hypothesized trend (the regional trend estimate is actually slightly biased). We then find the probability that the estimate lies within $0.2\bar{\beta}$ of $\bar{\beta}$ using normal probabilities.

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Table 1: RADM predicted regional average reductions in sulfate deposition by 2010

Northeast	32%
Mid Atlantic	29%
Appalachians	35%
Midwest	30%
South	30%
Plains	19%
Mississippi Delta	14%

Table 2: Regional variance totals $\times 10000$ and sum of 40 largest rectangle variances for 1987 stations, after 10 stations deleted using regional and rectangle variance criteria, and after 10 stations added using regional and rectangle criteria.

		deletion criterion		addition criterion	
	1987	reg.	rect.	reg.	rect.
Northeast	6.56	6.58	6.66	5.79	6.26
MidAtlantic	9.51	9.52	9.51	6.11	7.01
Appalachian	5.05	5.08	5.07	5.02	5.03
Midwest	4.53	4.69	4.81	4.53	4.53
South	5.80	5.81	5.80	5.79	5.13
Total eastern regional	31.46	31.69	31.86	27.24	27.96
Total rectangle	0.2081	0.2082	0.2081	0.1913	0.1683

Figure 1: Eleven Regions

Figure 2: Shape of Temporal Change

Figure 3: Locations of 1982-1986 stations ($\times$) and of all stations in operation at end of 1987 ($+$).

Figure 4: Standard errors of estimated decrease in 2010 of log sulfate concentration for data years 1995 through 2010 in Mississippi Delta, Middle Atlantic, Northeast, South, Appalachian, Midwest, and Plains regions (top to bottom).

Figure 5: Probability of estimated decrease in 2010 of log sulfate concentration being within 20% of modeled decrease for data years 1995 through 2010 in Appalachian, Northeast, Midwest, South, Middle Atlantic, Plains, and Mississippi Delta regions (top to bottom).

Figure 6: Probability of estimated decrease in 2010 of log sulfate concentration being within 20% of modeled decrease in data year 2000 for Northeast, Middle Atlantic, Appalachian, Midwest, South, Plains, and Mississippi Delta regions using modified continental variance estimates. Upper, center, and lower values for each region correspond to σ_L^2 set to zero, the value estimated from the data, and twice the value estimated from the data.

Figure 7: Standard errors of estimated decrease in 2010 of log precipitation-adjusted sulfate concentration for data years 1995 through 2010 in Mississippi Delta, Middle Atlantic, Northeast, South, Appalachian, Midwest, and Plains regions (top to bottom).

Figure 8: Probability of estimated decrease in 2010 of log precipitation-adjusted sulfate concentration being within 20% of modeled decrease for data years 1995 through 2010 in Appalachian, Midwest, Northeast, South, Middle Atlantic, Plains, and Mississippi Delta regions (top to bottom).

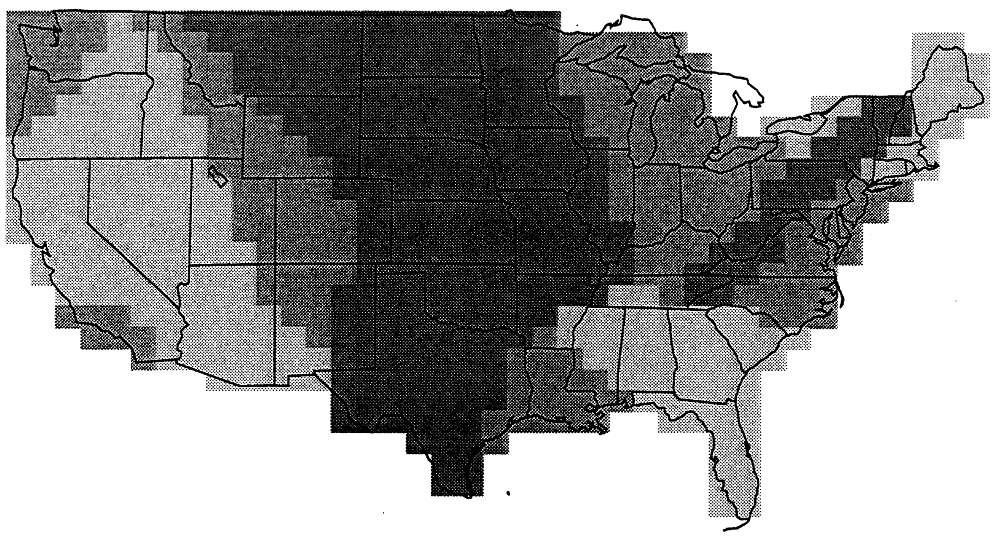
Figure 9: Locations of 40 largest rectangle variances for decrease in 2010 of log precipitation-adjusted sulfate concentration using all stations.

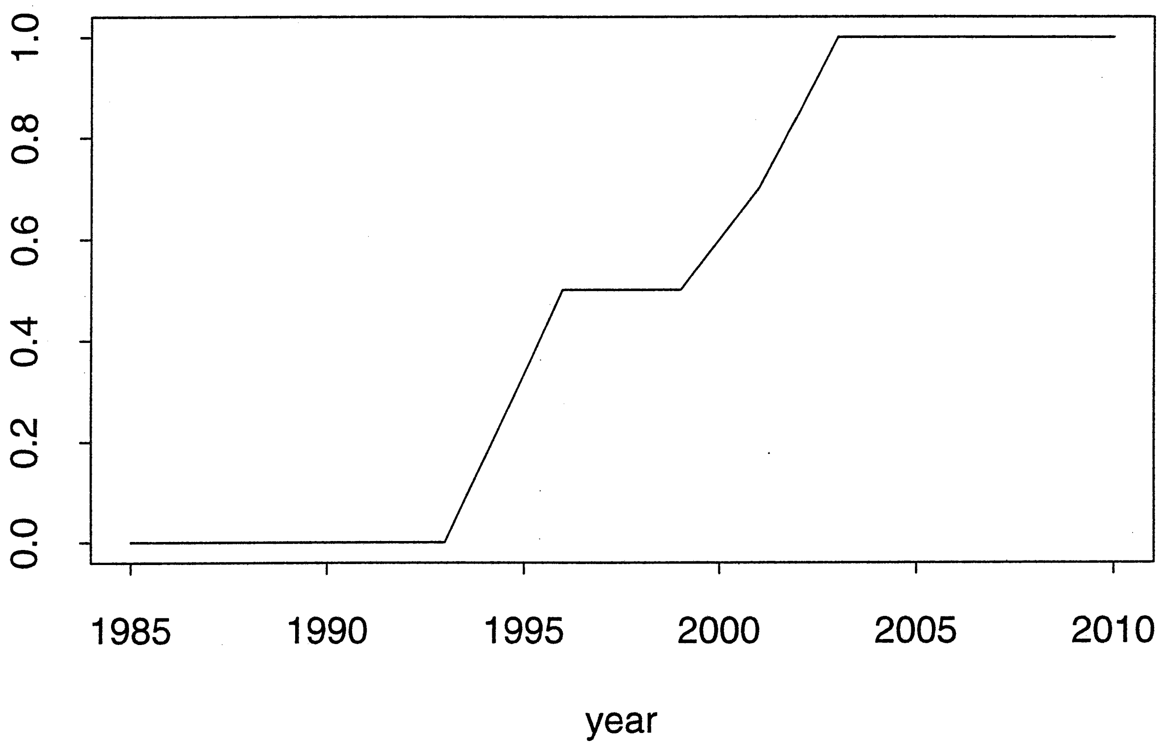
Figure 10: Locations of 40 largest rectangle variances for decrease in 2010 of log precipitation-adjusted sulfate concentration and deleted stations selecting on the regional variance criterion.

Figure 11: Locations of 40 largest rectangle variances for decrease in 2010 of log precipitation-adjusted sulfate concentration and deleted stations selecting on the rectangle variance criterion.

Figure 12: Locations of 40 largest rectangle variances for decrease in 2010 of log precipitation-adjusted sulfate concentration and added stations selecting on the regional variance criterion.

Figure 13: Locations of 40 largest rectangle variances for decrease in 2010 of log precipitation-adjusted sulfate concentration and added stations selecting on the rectangle variance criterion.

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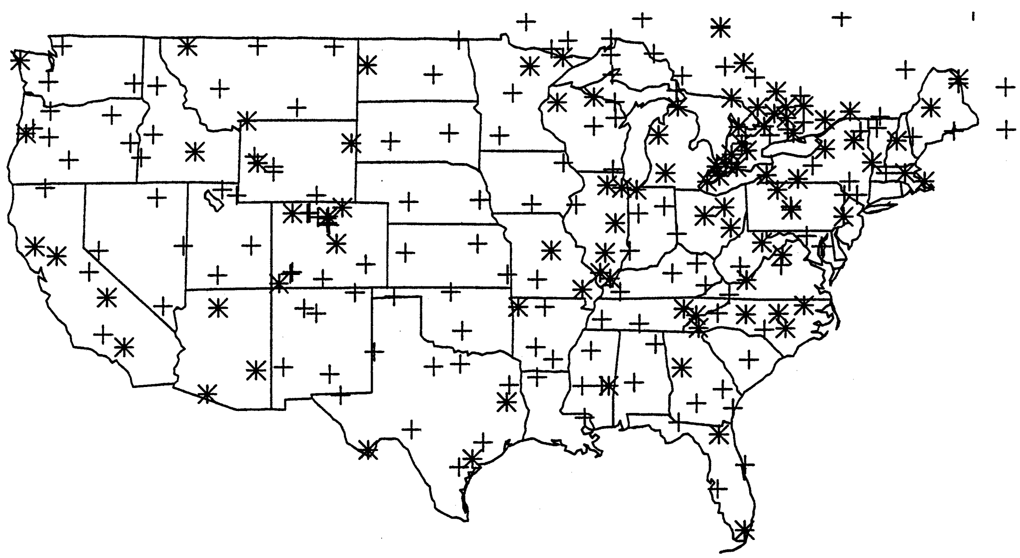

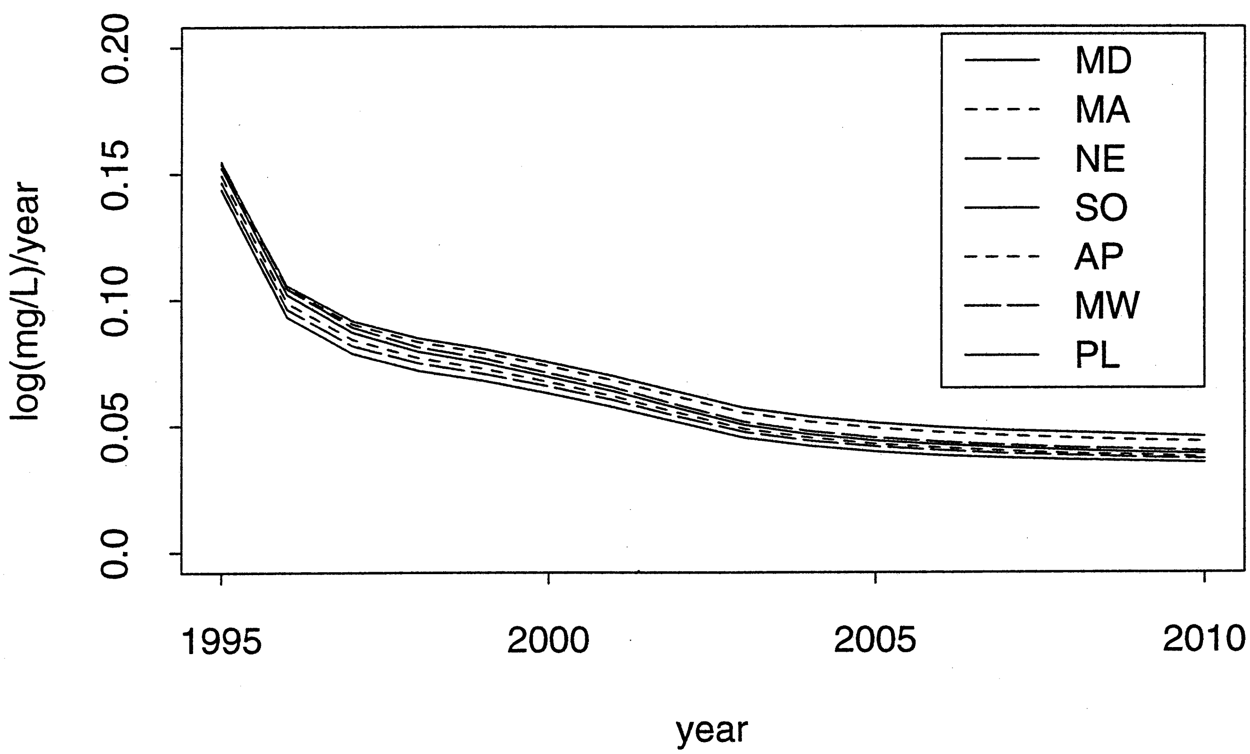

0.20
0.15
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log(mg/L)/year
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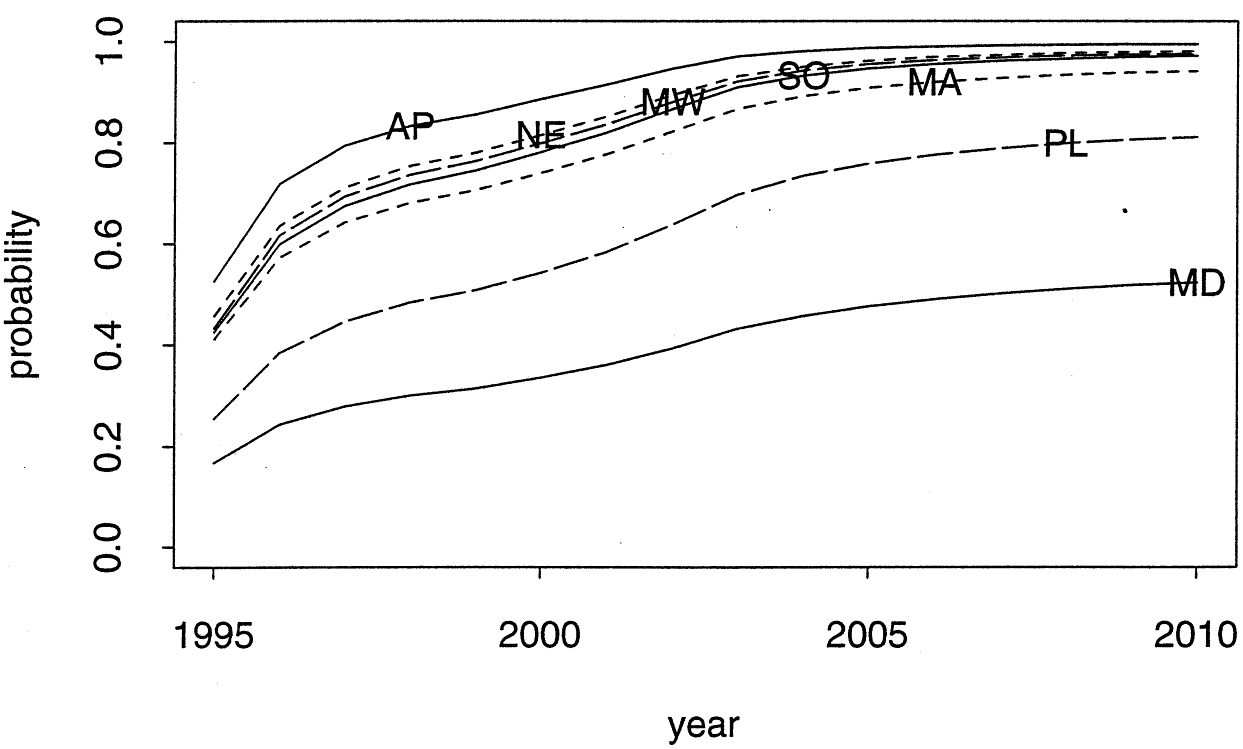

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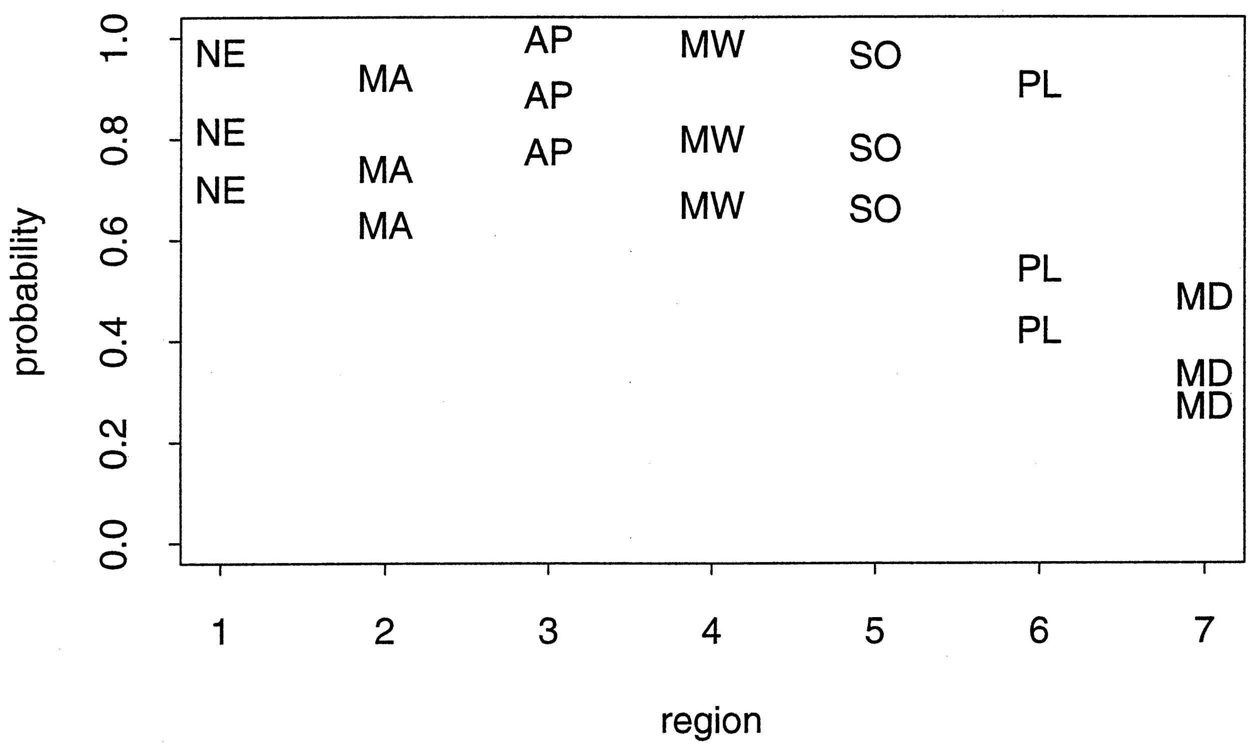
year



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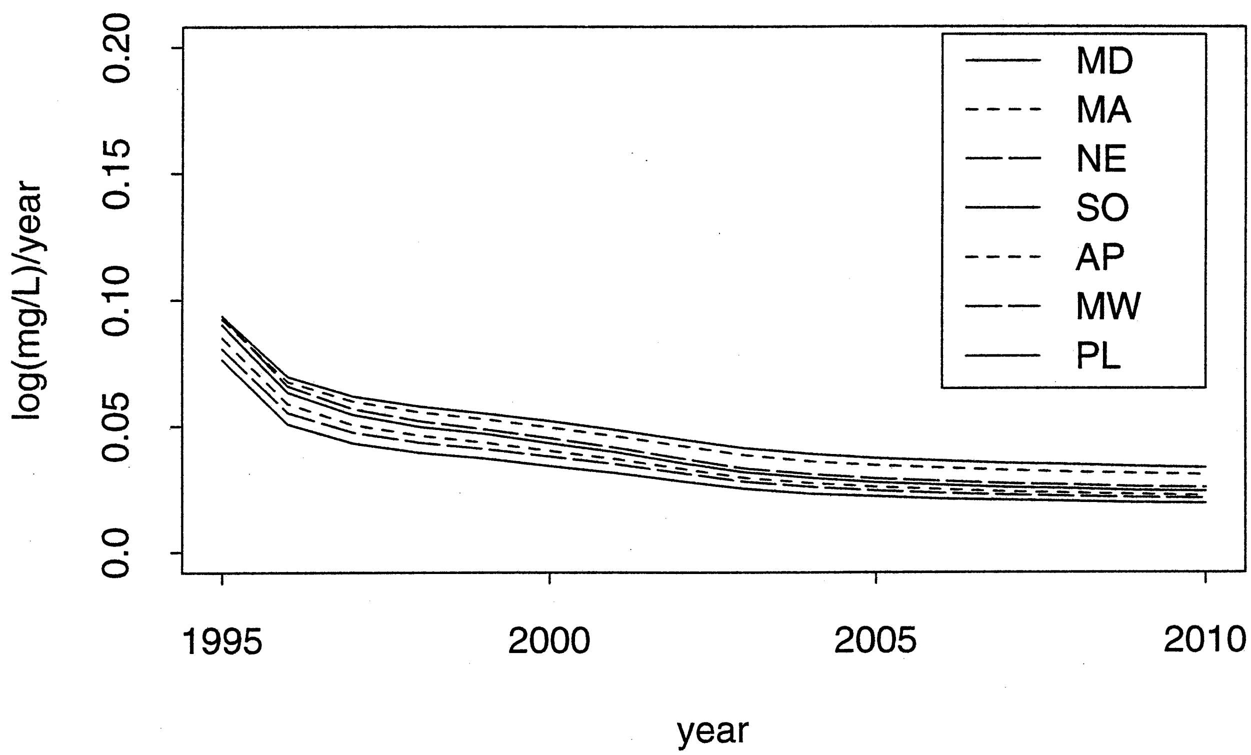

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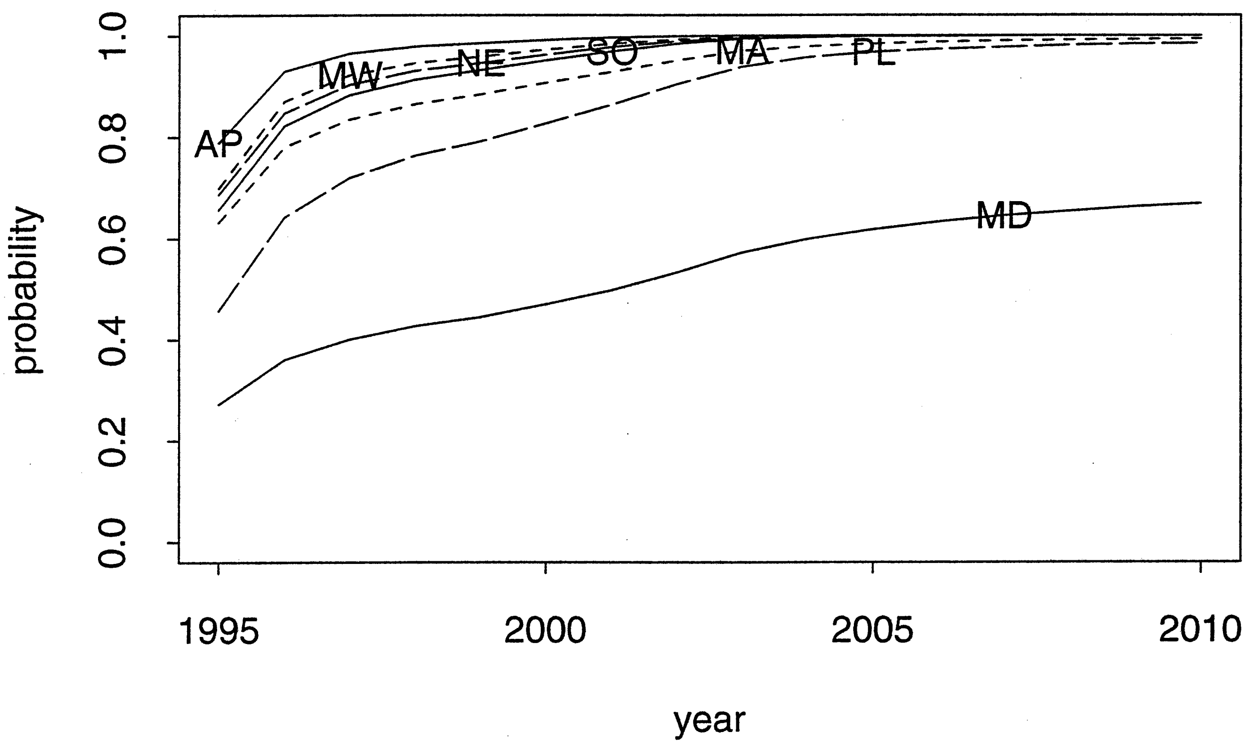
log(mg/L)/year
0.0
0.05
0.10
0.15
0.20
1995
2000
2005
2010
year



1.0
0.8
0.6
0.4
0.2
0.0
probability
AP
MW
NE
SO
MA
PL
MD
1995
2000
2005
2010
year