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AN INVESTIGATION OF CHANGES IN AIR PARTICULATE CONCENTRATIONS IN CHICAGO

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ABSTRACT

Changes in monthly averages of total suspended particulates in Chicago are analyzed to investigate the effect of a law forbidding the use of high sulphur coal, which came into effect in January 1970. A seasonal integrated autoregressive-moving average time series model is fitted to the data collected prior to 1970 and forecasts made from this model are compared with the data observed after the ban was in effect. A portmanteau test based on one-month ahead forecast errors indicates the decrease in particulate concentrations caused by the law banning the use of high sulphur coal to be statistically significant. Weight functions which represent the effects of changes in parameters of the model and changes in the series due to different factors modeled by a series of indicator variables are computed next. These weight functions are used to analyze the one-step ahead forecast error variance in order to evaluate the magnitude of changes in the long-term trend and seasonality of the particulate data. The results indicate a gradual decrease in the particulate emissions due to high sulphur coal burning that approaches steady-state after an approximate lag of five years.

I. INTRODUCTION

After a law which prohibited burning coal of high sulphur content came into effect in the city of Chicago, Illinois, U.S.A., the measured particulate concentration showed a drastic decrease in level as well as a substantial change in its characteristic periodic behavior. O'Neill analyzed the change in particulate concentration in Chicago by using an autoregressive model [1]. Rao and Padmanabhan used intervention analysis to investigate the changes in particulate

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concentration in warm and cold seasons after burning high sulphur coal was banned in Chicago [2].

There are some questions left unanswered by these studies. For example, the sources of variation of the level and periodicity of the particulate concentration series cannot be inferred by methods based on intervention analysis. A method developed recently by Box and Tiao [3], makes it possible to investigate the sources of variation in particulate concentration levels and also the changes in trend and seasonal characteristics of the time series after the ban on burning high sulphur coal went into effect. The basic objective of research presented herein is to investigate the sources of changes and the changes in seasonal behavior of the particulate concentration time series originated by the ban on burning high sulphur coal in Chicago by using Box and Tiao's method.

A time series of monthly average concentrations of air suspended particulates measured from January 1964 to December 1977 is used in the present study. The procedure used in the study is to analyze the errors in one-step ahead forecasts computed by means of a time series model based on data prior to the change in the series. The particle concentration data before January 1970 – i.e., before the ban on burning high sulphur coal went into effect – are used to construct and validate a time series model. This model is used to obtain one-month ahead forecasts of particle concentrations from January 1970 to December 1977. The resulting forecast errors are analyzed to test the changes in stochastic character of the time series after the law went into effect, and also to determine the sources of these changes. The sum of squares of one-month ahead forecast errors is compared to confidence intervals based on its theoretical distribution according to the model constructed by using data prior to the ban. The variations in computed forecast errors due to unit changes in model parameters and due to indicator variables are also investigated. The contributions of each of these factors to the sum of squares of errors are estimated via regression analysis.

This article is organized as follows. The particulate concentration data series and the stochastic model construction and validation for the data before January 1970 are discussed in section II. Testing the sum of squares of errors is discussed in section III. The computation of weight functions is discussed in section IV. The regression analysis of the forecast errors in the Chicago particulate data is discussed in section V. The results are discussed and a set of conclusions are presented in the last section.

II. DATA USED IN THE STUDY AND INITIAL TIME SERIES MODEL

The series of monthly averages of total suspended particulates measured at the central city monitoring location in Chicago is plotted, together with forecasts made with origin at the 72nd point ($t_0 = 72$) up to ninety-six months ahead

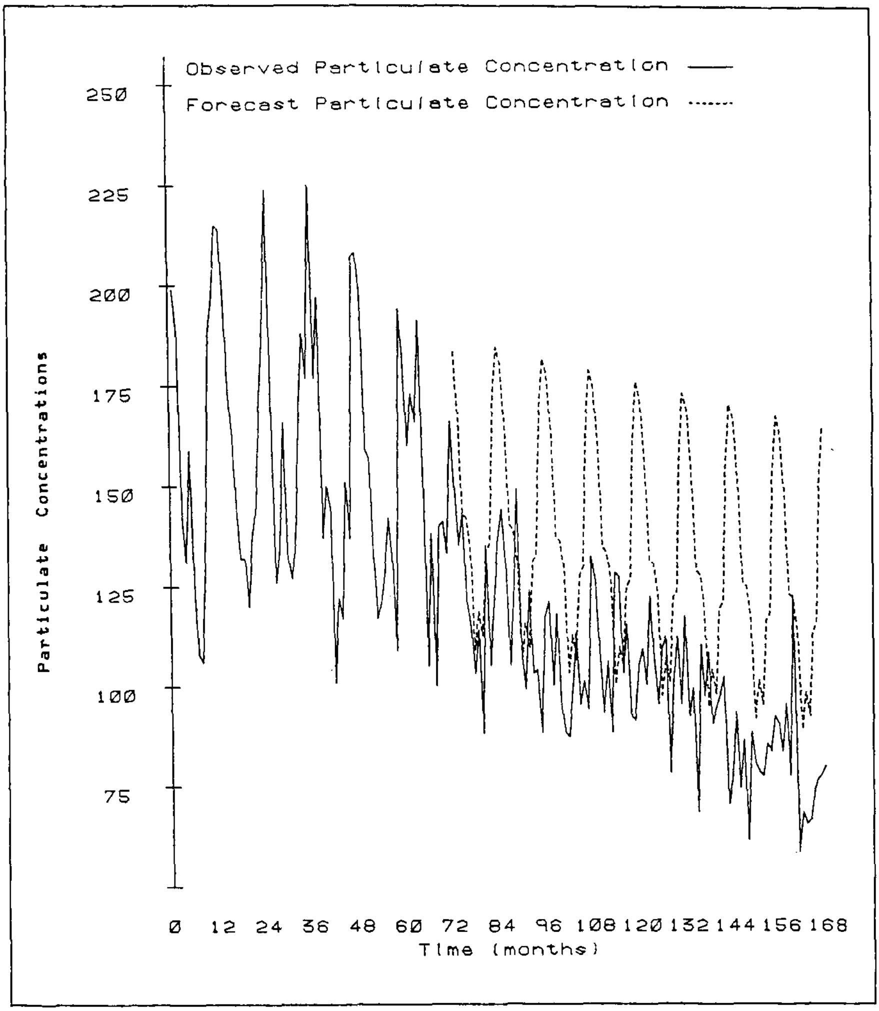


Figure 1. Chicago particulate time series and forecasts from equation 4 with origin at t = 72.

(l = 96), in Figure 1. The seventy-two data points observed before the ban came into effect, hereinafter referred to as the first part of the series, have a mean of 155.1, variance 1042, and skewness coefficient 0.364. This series is denoted by the sequence $z_1, z_2, \ldots, z_{t-1}, z_t, z_{t+1}, \ldots$. The monthly average particulate concentration z_t can be estimated by using a time series model which may involve past values $z_{t-1}, z_{t-2}, \ldots$. By using a white noise series, $\ldots a_{t-1}, a_t, a_{t+1}, \ldots$, defined as a sequence of independent identically distributed random variables, such a model can be described by the autoregressive (AR) model in equation (1),

$$a_t = \pi(B)z_t = (1 - \pi_1 B - \pi_2 B^2 - \ldots)z_t = z_t - \pi_1 z_{t-1} - \pi_2 z_{t-2} \ldots, \quad (1)$$

where B is the back shift operator such that $Bz_t = z_{t-1}$ and a_t is a normally distributed white noise series with mean zero and variance σ_a^2. O'Neill showed that twelve π-weights were adequate to model the Chicago particulate time series [1]. Alternatively, the moving average (MA) model equation (2) may also be used.

$$z_t = \psi(B)a_t = (1 + \psi_1 B + \psi_2 B^2 + \ldots)a_t = a_t + \psi_1 a_{t-1} + \psi_2 a_{t-2} \ldots. \quad (2)$$

Integrated autoregressive-moving average (ARIMA) models are usually more parsimonious than AR or MA models [4]. The general ARIMA model is represented by the finite difference equation (3)

$$\Phi(B^s)\phi(B)(1 - B)^d(1 - B^s)^D z_t = \theta_0 + \Theta(B^s)\theta(B)a_t . \quad (3)$$

This equation consists of the overall constant θ_0 and the following operators:

Regular Differencing	$(1 - B)^d$
Seasonal Differencing	$(1 - B^s)^D$
Regular Autoregressive (AR)	$\phi(B) = 1 - \sum_{i=1}^{p} \phi_i B^i$
Seasonal Autoregressive (SAR)	$\Phi(B) = 1 - \sum_{i=1}^{P} \Phi_i B^{si}$
Regular Moving Average (MA)	$\theta(B) = 1 - \sum_{i=1}^{q} \theta_i B^i$
Seasonal Moving Average (SMA)	$\Theta(B^s) = 1 - \sum_{i=1}^{Q} \Theta_i B^{si}$

In the present study a model with one seasonal differencing of order s = 12 is fitted to the first part of the series. After substituting the estimates of the overall constant θ_0 and the SMA parameter Θ_1 in (3), the model in equation (4) is obtained.

$$(1 - B^{12})z_t = \underset{(0.924)}{-2.946} + a_t - \underset{(0.159)}{0.791} a_{t-12} \quad (4)$$

The standard errors of the estimated parameters θ_0 and Θ_1 are given in parentheses below their respective values in (4). The residuals a_t are obtained by an iterative procedure which combines forecasting and backforecasting [4]. This procedure gives an estimated white noise variance $\hat{\sigma}_a^2 = 412$. The computed residuals are plotted in Figure 2 and their correlogram is shown in Figure 3. These residuals are tested for whiteness by using a portmanteau test. The details of this test are found in Box and Jenkins [4], and hence are not repeated here.

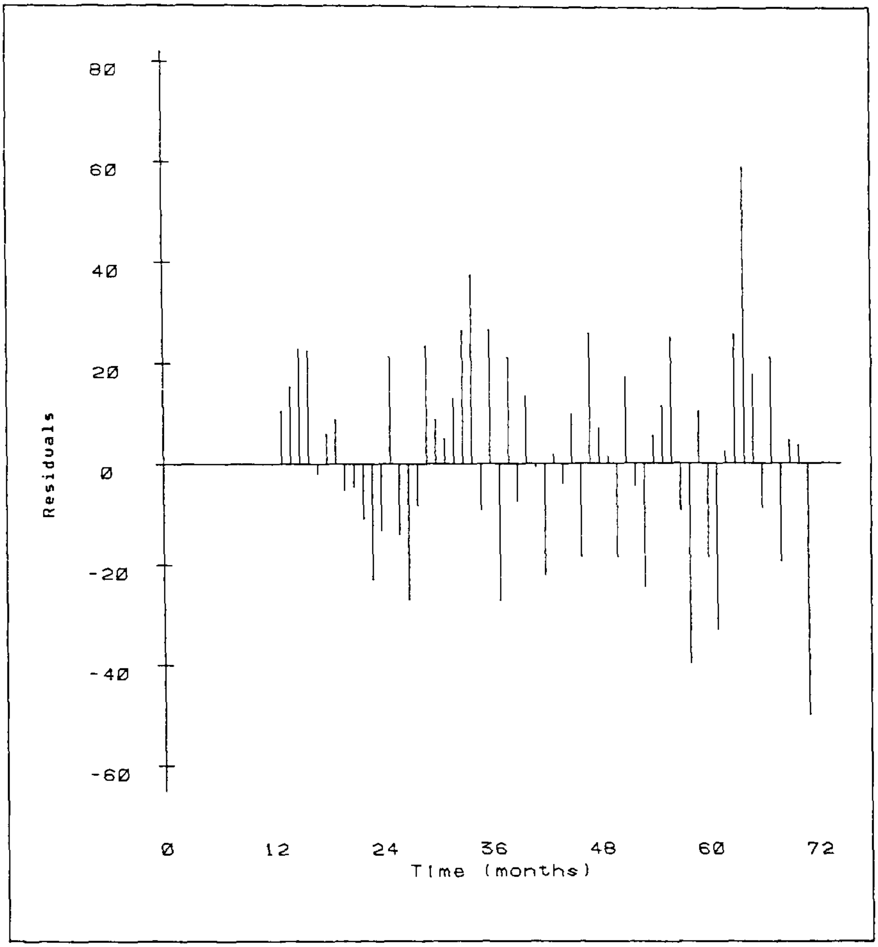


Figure 2. Residuals from the model fitted to the data before the ban (equation 4).

The value of the portmanteau statistic is Q = 11.6 which is well below the $\chi^2(0.95;13) = 22.4$ bound. Thus, the model in (4) is accepted as valid for the first part of the series. This model represents adequately the seasonality of the particulate concentrations due to household and industrial coal burning for heating and production purposes exhibited as a dominant feature in the first part of the series, as well as the weak decreasing trend present in the data.

The plot of the ninety-six observed particulate concentration values after seventy-two months, hereinafter referred to as the second part of the series (Figure 1), shows two major differences with respect to forecasts made from the

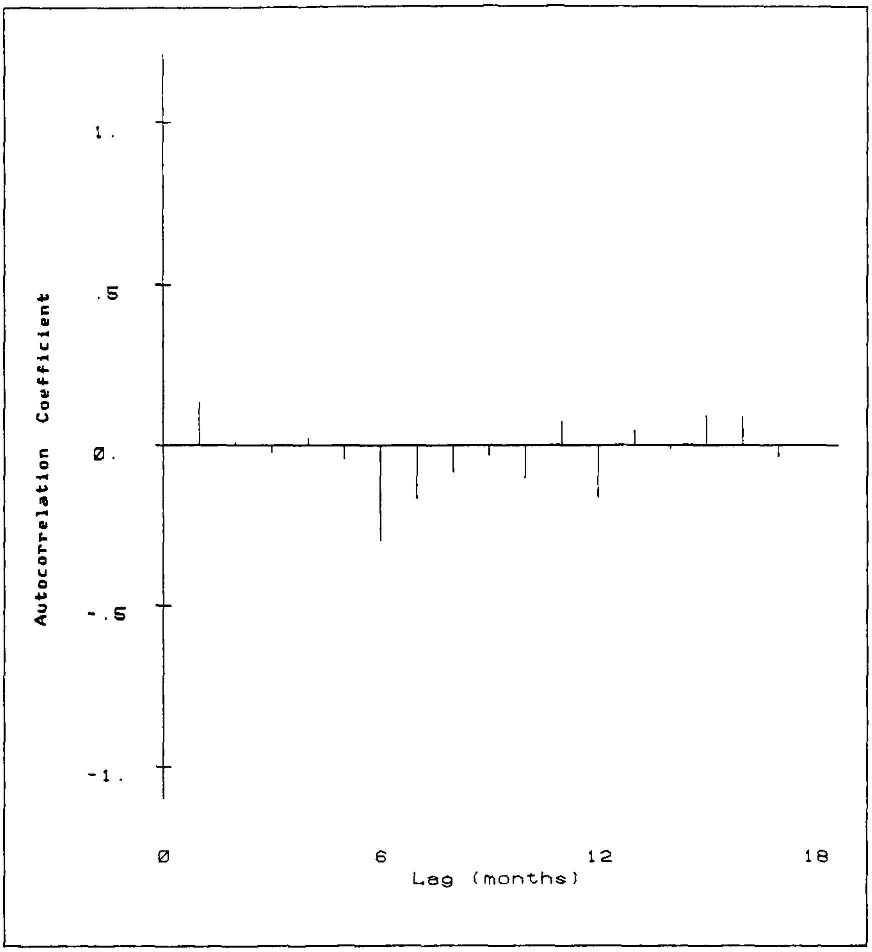


Figure 3. Correlogram of residuals from the model fitted to the data before the ban (equation 4).

model constructed for the first part of the series (equation 4). First, the slope of the trend line slants down. Secondly, the amplitude of the seasonal wave pattern has definitely decreased. A model similar in structure to the previous one is fitted to the second part of the series. Substitution of the corresponding parameter estimates gives model equation (5).

$$(1 - B^{12})z_t = -6.163 + a_t - 0.857a_{t-12} \qquad (5)$$
$$\qquad (0.437) \qquad (0.101)$$

Comparing equations (4) and (5), a change of approximately three negative units in θ_0 can be inferred, whereas the estimate of Θ_1 is not significantly different from that of the first part. This independent model is only an approximation since the probability distribution of the second part of the series is conditional on the observations and residuals from the first part of the data. In addition, this model does not account for the apparent change in the seasonal pattern of the data. These changes in the stochastic character of the series must therefore be analyzed by other methods such as those discussed in the next section.

III. TESTING THE FORECAST ERRORS

When the expected values $E[a_{t_0+1}] = 0$ for lead times $l = 1$ to $l = 96$ are substituted in (4) without updating the resulting forecasts with new observations, the plot of the observed and forecasted particulate concentrations for the second part of the series (Figure 1) give an impression of large discrepancy. However, this impression may be misleading since the effect of assuming the one-step ahead forecast errors to be zero is cumulative. In order to test the significance of this discrepancy, it is first assumed that the parameters of the model equation 4 have not changed and the one-step ahead forecast errors for the second part, denoted here by $\hat{a}'_t$ are computed by using (6).

$$\hat{a}'_t = \hat{\pi}(B)z_t. \tag{6}$$

When the ARIMA model difference equation (3) is preferred, (6) is written as

$$\hat{\Phi}(B^s)\,\hat{\phi}(B)(1 - B)^d(1 - B^s)^D z_t = \hat{\theta}_0 + \hat{\Theta}(B^s)\hat{\theta}(B)\hat{a}'_t\,. \tag{7}$$

After expanding (7), it can be solved recursively for $\hat{a}'_t$ in terms of the observed series $z_t, z_{t-1}, \ldots, z_{t-d-sD-p-sP}$, and computed residuals $\hat{a}'_{t-1}, \hat{a}'_{t-2}, \ldots, \hat{a}'_{t-q-sQ}$. The recursive computation starts by substituting the previously computed residuals $\hat{a}'_t$ from the first part instead of the corresponding $\hat{a}'_t$ for $t < t_0$.

When the pertinent parameter estimates from (4) are substituted in (7), the one-step ahead forecast errors for the Chicago data are computed by using (8).

$$\hat{a}'_t \;=\; z_t - z_{t-12} + 2.946 + 0.791\,\hat{a}'_{t-12}. \tag{8}$$

If the ban on burning high sulphur coal has brought about no change in the time series, the forecast errors given by equation (8) would constitute a white noise series and their standardized sum of squares Q_1 during the period $t_0 + 1, \ldots, t_0 + 1$,

$$Q_1 = \frac{\sum_{i=1}^{1} (\hat{a}'_{t_0+1})^2}{\hat{\sigma}_a^2} \tag{9}$$

would be distributed as χ^2 with 1 degrees of freedom. Thus, a *portmanteau* test of the continuing appropriateness of the model during this period is achieved by comparing Q_1 with the corresponding χ^2 bound for a given confidence level. If the value of Q_l is above this bound, then we may conclude that the stochastic character of the time series has changed.

The one-step ahead forecast errors of the second part of the Chicago particulate series, plotted in Figure 4, show an apparent lack of randomness. By using the estimated residual variance from the model fitted to the first part of the series ($\hat{\sigma}_a^2 = 412$), Q_{96} is calculated as 161.6, which is much larger than $\chi^2(0.95;96) = 119.9$. The Q_l values for lead times $l = 1$ to $l = 96$ are plotted on Figure 5. These Q_l values are highly above the $\chi^2(0.95;l)$ bounds after two years since the ban went into effect. Thus, the stochastic character of the

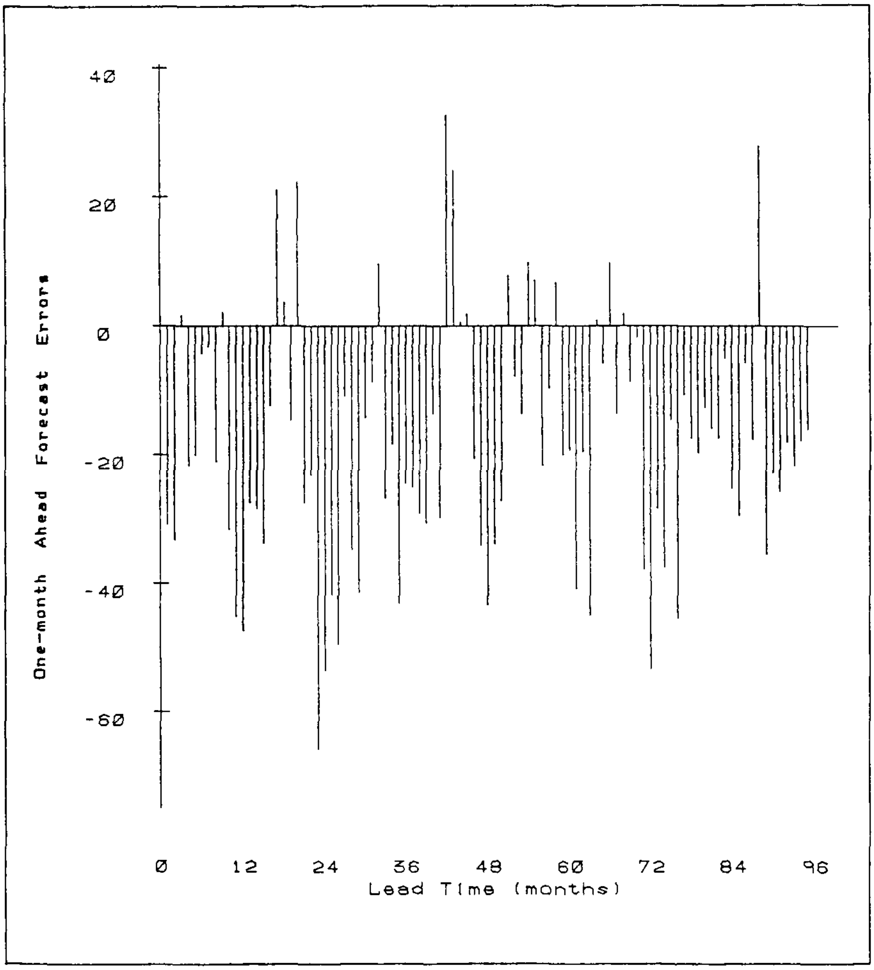


Figure 4. One-month ahead forecast errors after the ban was in effect computed by using equation 8.

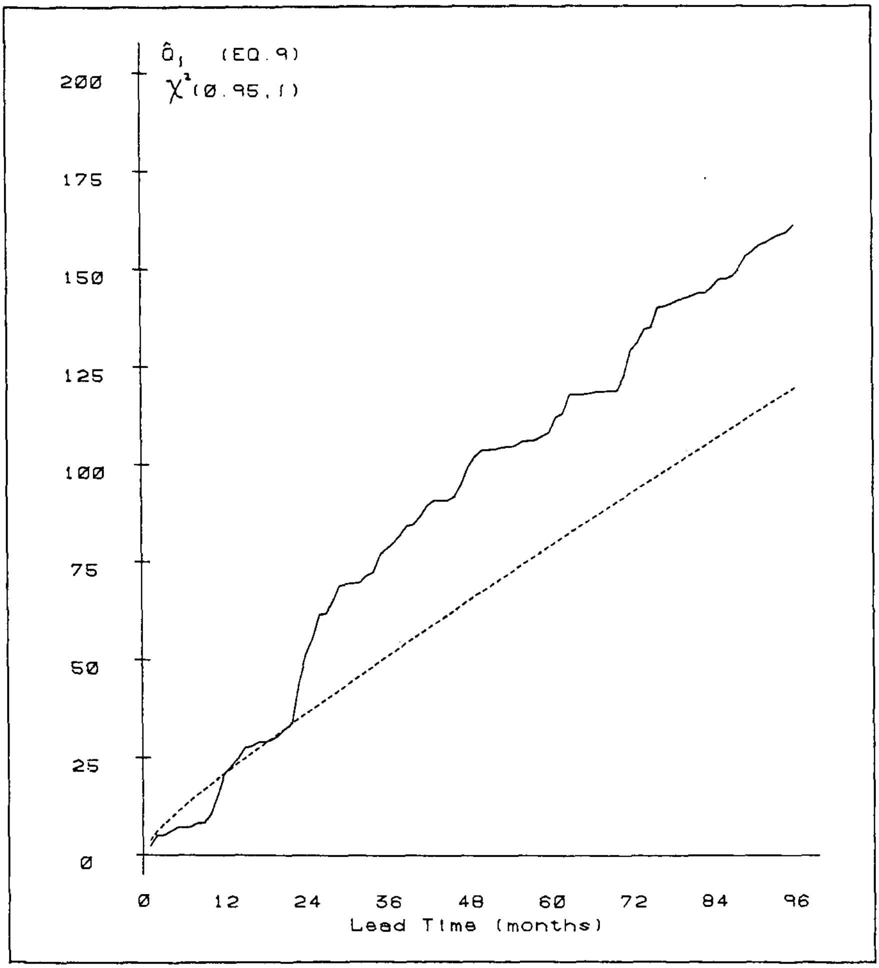


Figure 5. Portmanteau statistic of one-month ahead forecast errors from the model fitted to the data before the ban.

particulate concentration series has been definitely altered after the law banning the use of high sulphur coal.

IV. COMPUTATION OF CHANGE WEIGHT FUNCTIONS

The changes which have been introduced into the system may be represented by the model equation (10).

$$\pi'(B)z_t' = a_t. \tag{10}$$

Let the operator $\pi(B)$ of equation (1) be expressed in terms of n parameters. Each of these parameters, denoted here by γ_k, is incremented by an amount $\Delta\gamma_k$ and the resulting values $\gamma_k' = \gamma_k + \Delta\gamma_k$ are substituted into the operator $\pi'(B)$ of equation (10). Let the series z_t' be derived by equation (11).

$$z_t' = z_t - \sum_{j=1}^{m} \beta_j x_{jt} , \tag{11}$$

where x_{jt} are m series of indicator variables representing different agents which may be responsible for changes in the original series. Several types of functions may be used to represent step changes, seasonal changes, or the influence of new exogenous variables. The coefficients β_j are quantitative measures of the influence of the corresponding series of indicator variables. Considering these combined changes, the one-step ahead forecast errors $\hat{a}_t'$ can be expressed in terms of n weight function series $W_{\gamma_k t}$ for the changes in parameters $\Delta\gamma_k$ and m series X_{jt} for the effects of indicator variables β_j as shown in (12) [5].

$$\hat{a}_t' = \sum_{j=1}^{m} \beta_j X_{jt} + \sum_{k=1}^{n} \Delta\gamma_k W_{\gamma_k t} + a_t , \tag{12}$$

where the X_{jt} and $W_{\gamma_k t}$ weights are computed as follows:

Change in Parameter γ_k $\qquad W_{\gamma_k t} = - \dfrac{\partial \hat{a}_t'}{\partial \gamma_k}$ (13)

Indicator Series x_{jt} $\qquad X_{jt} = \pi(B) x_{jt}$ (14)

A brief derivation of equations (12), (13), and (14) from (6), (10), and (11), is presented in Appendix A.

When the model fitted to the first part of the data is given in the form of the ARIMA difference equation (3), the one-step ahead forecast errors of the second part of the series are computed by using (7). Then, (14) is written as (15a).

$$\hat{\Phi}(B^s)\hat{\phi}(B)(1 - B)^d(1 - B^s)^D x_{jt} = \hat{\Theta}(B^s)\hat{\theta}(B)X_{jt} . \tag{15a}$$

Thus, the computation of the series of weights X_{jt} is equivalent to substituting such weights for the one-step ahead forecast errors $\hat{a}_t'$ when the time series of indicator variables x_{jt} is substituted for z_t in (7). Since the term $\hat{\theta}_0$ has been already accounted for in (7), it does not affect the indicator series in (15a).

By partial differentiation of (7) with respect to each parameter, equation (13) is written in the forms given in Table 1. Thus, the series of weights $W_{\gamma_k t}$ can be obtained as one-step ahead forecast errors from the observed series z_t or computed $\hat{a}_t'$ with respect to appropriate modifications of the model equation (7). Equations (15) can be solved recursively for the X_{jt} and $W_{\gamma_k t}$ series in a similar fashion as (7). Since the model parameters are constant before the introduced changes, the starting values $W_{\gamma_k t}$ as well as X_{jt} are zeros for $t \leqslant t_0$.

Table 1. Equations for Computation of Weight Functions

Parameter	*Equation for the W-Weights*	*Equation Number*
ϕ_i	$\hat{\Phi}(B^s)(1-B)^d(1-B^s)^D B^i z_t = \hat{\Theta}(B^s)\hat{\theta}(B)W_{\phi_i t}$	(15b)
Φ_i	$\hat{\phi}(B)(1-B)^d(1-B^s)^D B^{is} z_t = \hat{\Theta}(B^s)\hat{\theta}(B)W_{\Phi_i t}$	(15c)
θ_i	$B^i \hat{a}_t' = -\hat{\theta}(B)W_{\theta_i t}$	(15d)
Θ_i	$B^{is}\hat{a}_t' = -\hat{\Theta}(B^s)W_{\Theta_i t}$	(15e)
θ_0	$1 = \hat{\Theta}(B^s)\hat{\theta}(B)W_{\theta_0 t}$	(15f)

Weight Series for the Chicago Particulate Data

For the Chicago particulate data, changes in parameters θ_0 and Θ_1 are investigated. In addition, the series $\hat{a}_t'$; $t > t_0$, is partitioned in several subsets whose correlograms (Figure 6) clearly indicate that these residuals have a strong seasonal component which decreases until vanishing at approximately five years after the law was enacted. Thus, a series of indicator variables x_{1t} is included in the model to account for this "damping" of the seasonal component present in the first part of the series z_t. Substituting this damping component in (11), equation (16) is obtained.

$$z_t' = z_t - \beta_1 x_{1t}, \tag{16}$$

where β_1 represents the initial amplitude of the gradually decreasing seasonal component.

In order to obtain an adequate expression for x_{1t}, the coefficients of a Fourier series expansion for the seasonal component of the first part of series are first calculated. After the linear trend is deleted, the coefficients estimated by regression analysis of the data from the first period give a periodic function which is then rescaled by dividing by its amplitude. The resulting unit amplitude seasonal indicator function is given in (17a).

$$\begin{aligned} u_t = {} & 0.7550\cos\frac{\pi t}{6} + 0.2666\sin\frac{\pi t}{6} + 0.1804\cos\frac{\pi t}{3} \\ & -0.0298\sin\frac{\pi t}{3} + 0.0669\cos\frac{\pi t}{2} + 0.1076\sin\frac{\pi t}{2} \\ & -0.0441\cos\frac{2\pi t}{3} - 0.0191\sin\frac{2\pi t}{3} + 0.0840\cos\frac{5\pi t}{6} \\ & -0.1037\sin\frac{5\pi t}{6} - 0.0420\cos\pi t. \end{aligned} \tag{17a}$$

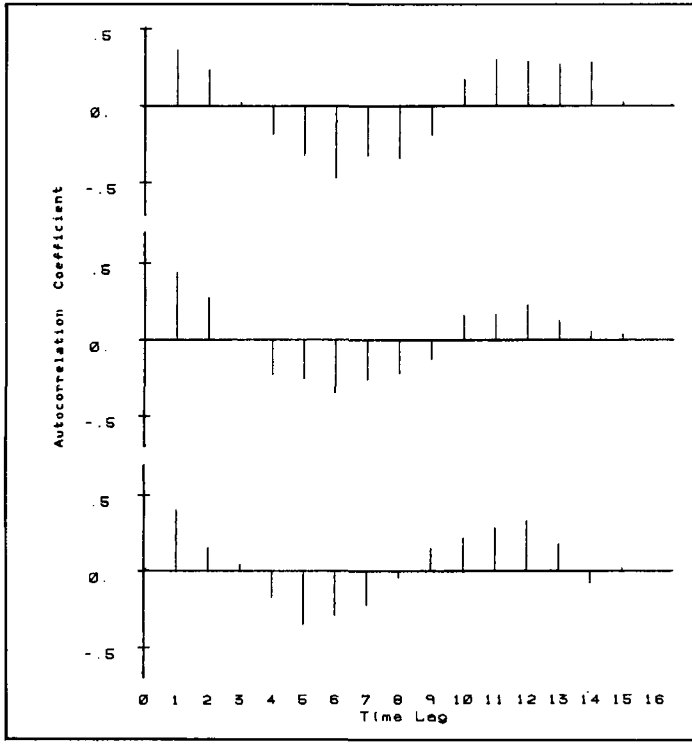
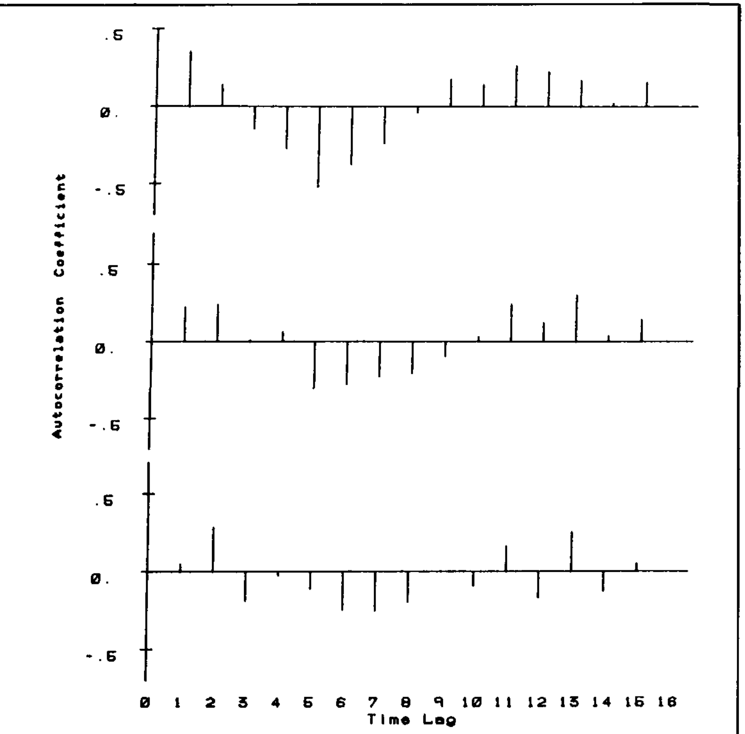


Figure 6. Correlograms of one-month ahead forecast errors after the ban was in effect. Top: years 1 to 3 (1970-1972); Center: years 2 to 4 (1971-1973); Bottom: years 3 to 5 (1972-1974).



Correlograms of one-month ahead forecast errors after the ban was in effect. Top: years 4 to 6 (1973-1975); Center: years 5 to 7(1974-1976); Bottom: years 6 to 8 (1975-1977).

The periodic function u_t is plotted in Figure 7a. The function y_t given by (17b) decreases parabolically from y = 0 at t = 72 to y = -1 at t = 132. This function y_t, shown in Figure 7b, represents a gradual decrease in amplitude during that period.

$$y_t = \begin{cases} 0 & 0 < t \leqslant 72 \\ \left(\frac{t-132}{60}\right)^2 - 1 & 72 < t \leqslant 132 \\ -1 & t > 132 \, . \end{cases} \qquad (17b)$$

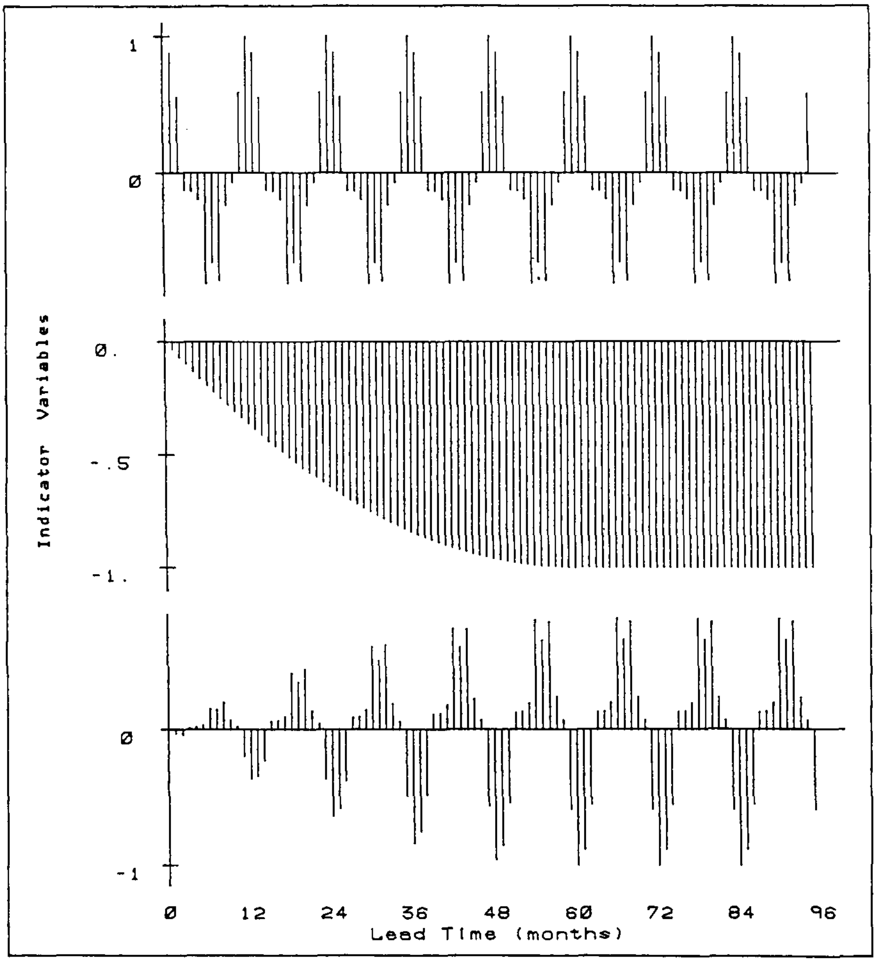


Figure 7. Construction of the series of change indicator variables. Top: unit amplitude seasonal indicator (eq. 17a); Center: parabolic decrease indicator (eq. 17b); Bottom: seasonality change indicator (eq. 17c).

The indicator series included in the model is the product of the functions y_t and u_t, given in (17c) and plotted in Figure 7c.

$$x_{1t} = u_t y_t \,. \tag{17c}$$

The weights for the change due to x_{1t} are obtained by substituting the parameter Θ_1 from model equation (4) into (15), yielding

$$(1 - B^{12})x_{1t} = (1 - 0.791B^{12})X_{1t}. \tag{18a}$$

Similarly, the weights for changes in the SMA parameter Θ_1 and the trend parameter θ_0 are computed by substituting Θ_1 into (15c) and (15f), yielding equations (18b) and (18c) respectively.

$$\hat{a}'_{t-12} = -(1 - 0.791B^{12})W_{\Theta_1 t} \tag{18b}$$

$$1 = (1 - 0.791B^{12})W_{\theta_0 t}. \tag{18c}$$

Equations (18) are solved for recursive computation of the weights as equations (19).

$$X_{1t} = x_{1t} - x_{1,t-12} + 0.791X_{1,t-12} \tag{19a}$$

$$W_{\Theta_1 t} = -\hat{a}'_{t-12} + 0.791W_{\Theta_1 t-12} \tag{19b}$$

$$W_{\theta_0 t} = 1 + 0.791W_{\theta_0 t-12} \tag{19c}$$

The resulting weights X_{1t}, $W_{\Theta_1 t}$, and $W_{\theta_0 t}$ are plotted in Figures 8a, 8b, and 8c, respectively. The contribution of each of these sources of variation to the total forecast errors is evaluated next.

V. RESULTS OF REGRESSION ANALYSIS OF THE FORECAST ERRORS

After computing the forecast errors $\hat{a}_t'$ and the series of weights X_{1t}, $W_{\Theta_1 t}$, and $W_{\theta_0 t}$, a regression equation is fitted to evaluate the contribution of each source to the total change in residual variance. By substituting the pertinent weights into (12), the amplitude of the damped seasonal component β_1 and the changes in parameters $\Delta\Theta_1$ and $\Delta\theta_0$ are expressed as regression coefficients in (20).

$$\hat{a}_t' = \beta_1 X_{1t} + \Delta\Theta_1 W_{\Theta_1 t} + \Delta\theta_0 W_{\theta_0 t}. \tag{20}$$

The coefficients of all combinations of 1, 2, and 3 non-zero regression terms in this equation are calculated in order to select the best model among them to explain the changes in the characteristics of the particulate data. The portion of the sum of squares due to regression (SSR), along with the corresponding regression coefficients for each combination, is shown in Table 2. The portion

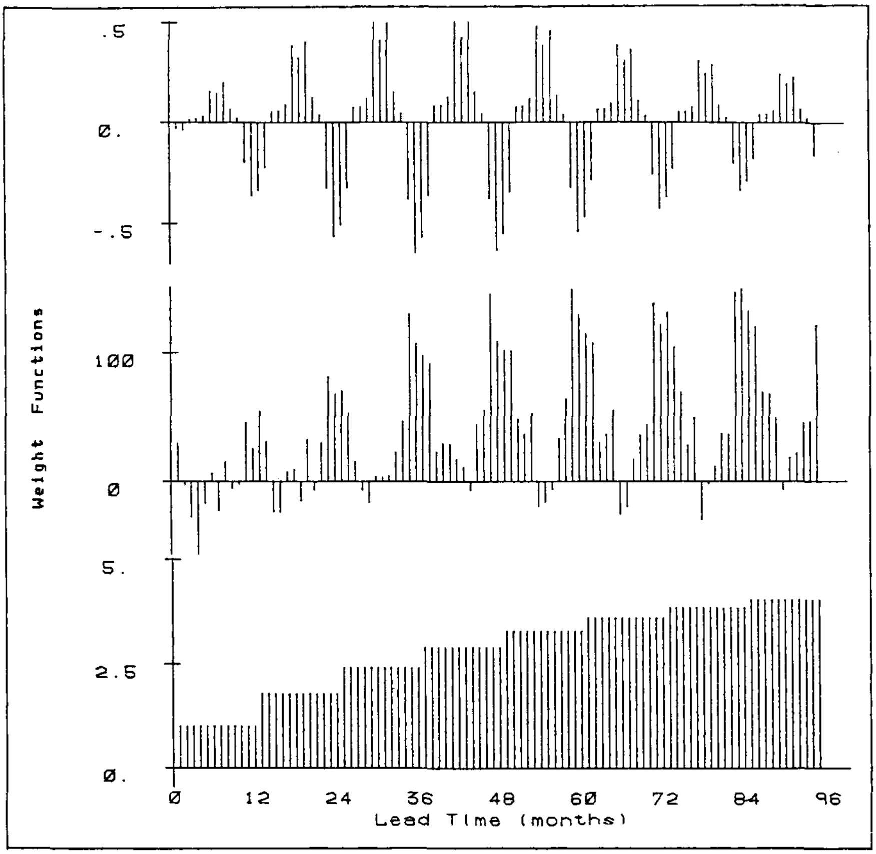


Figure 8. Weight functions for changes in the series. Top: change due to indicator variables (eq. 18a); Center: change in SMA parameter (eq. 18b); Bottom: change in MA constant (eq. 18c).

of the *portmanteau* statistic Q_{96} explained by each model is obtained by dividing the corresponding SSR by the previous estimated white noise variance $\hat{\sigma}_a^2$. The portion of Q_{96} due to the error sum of squares SSE, is then obtained by subtraction.

Selection of Best Regression Model

Model 7 gives a value $\hat{\Theta}_1' = 1.008$, which violates the conditions for stationarity of the system, whereas the Q_{96} due to SSE in model 3 is above the $\chi^2(0.95;96) = 119.9$ bound. Hence these two models are not considered further. The marginal contribution of each regression term in the remaining models from Table 2 is then obtained as the difference in Q_{96} due to SSR of models before and after including the term under consideration, as given in Table 3.

Table 2. Regression Analysis of One-Step Ahead Forecast Errors

Model Number	*Source of Variation*	*Regression Coefficients*	*SSR*	*Q_{96} due to SSR*	*Q_{96} due to SSE*
1	Change in θ_0 only	$\Delta\theta_0 = -5.525$	26972	65.6	96.0
2	Change in Θ_1 only	$\Delta\Theta_1 = -0.246$	26278	63.8	97.8
3	Change due to x_{1t} only	$\beta_1 = 40.71$	13285	32.3	129.3
4	Change in θ_0 and due to x_{1t}	$\Delta\theta_0 = -5.526$ $\beta_1 = 40.71$	40259	97.8	63.8
5	Change in θ_0 and Θ_1	$\Delta\theta_0 = -3.316$ $\Delta\Theta_1 = -0.138$	30919	75.1	86.5
6	Change in Θ_1 and due to x_{1t}	$\Delta\Theta_1 = -0.217$ $\beta_1 = 12.03$	27063	65.8	95.8
7	Change in θ_0, Θ_1, and due to x_{1t}	$\Delta\theta_0 = -9.020$ $\Delta\Theta_1 = 0.218$ $\beta_1 = 69.60$	43443	105.5	56.1

Table 3. Marginal Contribution of Regression Terms to χ^2 Statistic

Coefficient	*Model No.*	*Q_{96} due to SSR Before Inclusion*	*Q_{96} due to SSR After Inclusion*	*Marginal Q_{96}*
$\Delta\theta_0$	1	0	65.6	65.6
$\Delta\Theta_1$	2	0	63.8	63.8
β_1	4	65.6	97.8	32.2
$\Delta\Theta_1$	5	65.6	75.1	9.5
β_1	6	63.8	65.8	2.0

From these results the relative significance of each source of variation is inferred. Among one-term models, $\Delta\theta_0$ in model 1 has the largest contribution to the χ^2 value, yet this model does not account for change in the seasonal character of the particulate series. The contributions of $\Delta\Theta_1$ and β_1 in models 5 and 6 respectively, are very small. Model 4 has a large contribution of β_1 after initial inclusion of $\Delta\theta_0$. From these considerations, model 4 (equation 21) is selected as best among the models presented above.

$$\hat{a}_t' = \beta_1 X_{1t} + \Delta\theta_0 W_{\theta_0 t} + a_t . \tag{21}$$

Consequently, the changes in the Chicago particulate time series is best explained by a change in the slope of the trend line, represented by $\Delta\theta_0 = -5.526$, and a decrease in the amplitude of the seasonal wave pattern, represented by $\beta_1 = 40.71$.

An Alternative Model

A change in slope of the trend line of the Chicago particulates is represented in the model selected above by a change in the parameter θ_0. This change in slope can be also represented by an indicator series with value zero for $t \leqslant t_0$ and increasing linearly thereafter. O'Neill proposed a model with a parallel shift represented by a time series with value zero for $t < 72$ and one for $t \geqslant 72$ [1]. A model which represents a change in level between these two and also includes the change in the seasonal component of the series is developed from physical considerations, and the results of this model are presented herein.

As changes in the particulate concentration appeared after the ban on burning high sulphur coal went into effect, it is reasonable to assume that the subsequent reduction in high sulphur coal particle sources followed a "first order reaction law." More explicitly, the rate of reduction in the number of furnaces using high sulphur coal at a certain time is assumed to be proportional to the concurrent number of such furnaces. In constructing this model, the weak decreasing trend present in the first part of the series, represented by the parameter θ_0, is assumed to be due to other sources whose identification is beyond the scope of this study. Thus, considering that all conditions other than the emission of particulates due to burning high sulphur coal remain unchanged, the same linear trend can be assumed to be present in the series after this first order change has reached steady state.

In order to investigate this kind of change, the amplitude of the seasonal component of the series is first assumed to be reduced in annual steps [6]. Thus, eight indicator time series, representing the step reduction corresponding to each year after the ban, are constructed. Each of these series has a value of 0 before that year and -1 thereafter. When these series are substituted for y_t in (17c), eight time series of seasonal indicator variables $x_{1t}, \ldots, x_{8t}$, are obtained. The corresponding coefficients β_1 to β_8 are obtained by regression and then added up

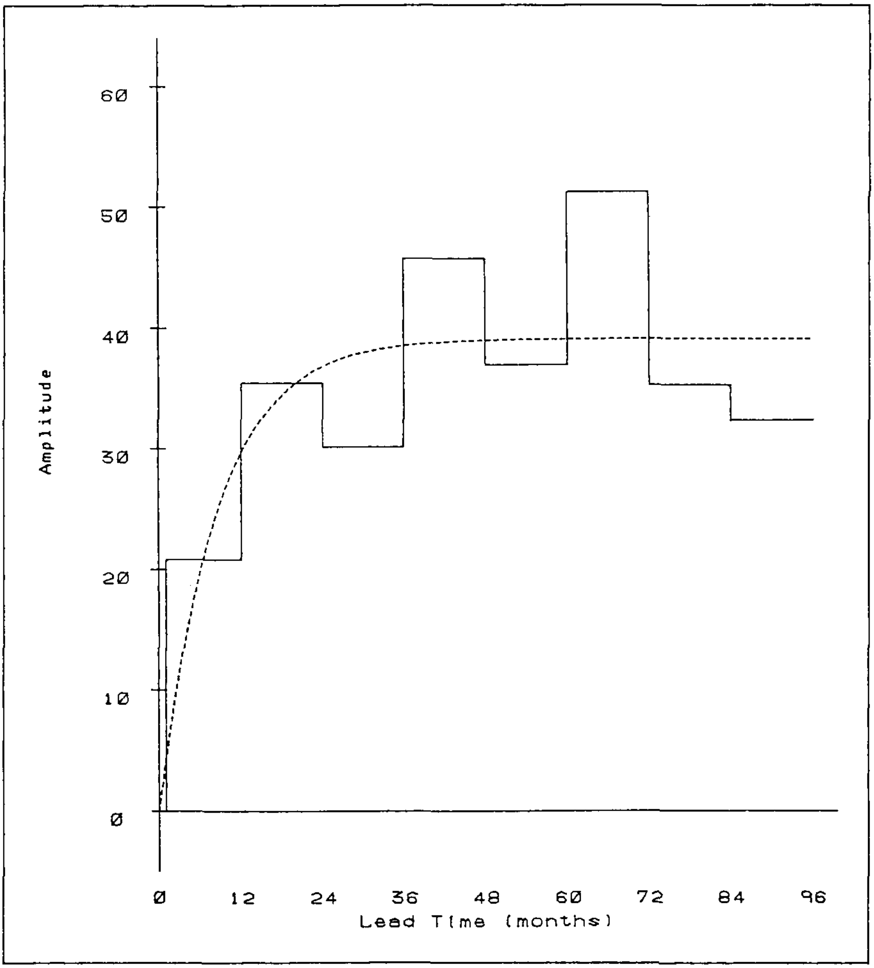


Figure 9. Cumulative step reduction in amplitude of the seasonal component.

to give the cumulative step change plotted against time in Figure 9, showing that the reduction in amplitude of the seasonal component can be adequately fitted by a first order reaction law of the same type as the time series represented in (21a) (the sign has been changed to indicate decrease).

$$x_{2t} = \begin{cases} 0 & 0 < t \leqslant 72 \\ e^{-\alpha(t-72)} - 1 & 72 < t \leqslant 132 \end{cases} \tag{21a}$$

Successive approximations given an optimal value of the exponent $\alpha = 0.0796$ and the coefficients of the unit amplitude seasonal series with a similar form as the series u_t from equation (17a), given as s_t in (21b)

Table 4. Regression Analysis of First Order Reaction Model

Model Number	*Source of Variation*	*Regression Coefficients*	*SSR*	*Q_{96} due to SSR*	*Q_{96} due to SSE*
1	Change due to x_{1t} only	$\beta_1 = 37.29$	14960	36.3	125.3
2	Change due to x_{2t} only	$\beta_2 = 35.07$	30129	73.2	88.4
3	Change due to x_{1t} and x_{2t}	$\beta_1 = 36.71$ $\beta_2 = 34.80$	44620	108.4	53.2

$$\begin{aligned} s_t = {} & 0.8233\cos\frac{\pi t}{6} + 0.2823\sin\frac{\pi t}{6} + 0.1534\cos\frac{\pi t}{3} \\ & -0.1374\sin\frac{\pi t}{3} + 0.0663\cos\frac{\pi t}{2} - 0.0036\sin\frac{\pi t}{2} \\ & -0.0540\cos\frac{2\pi t}{3} - 0.1329\sin\frac{2\pi t}{3} + 0.0901\cos\frac{5\pi t}{6} \\ & -0.1355\sin\frac{5\pi t}{6} - 0.0970\cos\pi t\,. \end{aligned} \tag{21b}$$

The product of s_t and x_{2t} gives an indicator series for the reduction in amplitude of the seasonal component expressed as x_{1t} in (21c).

$$x_{1t} = s_t x_{2t}\,. \tag{21c}$$

The complete model to explain the changes in particulate concentration is formulated in terms of the corresponding weights X_{1t} and X_{2t} as in (22),

$$\hat{a}_t' = \beta_1 X_{1t} + \beta_2 X_{2t} + a_t\,, \tag{22}$$

where β_1 is interpreted as in the model presented before (equation 21), and β_2 represents the reduction in the mean annual concentration of particulates from high sulphur coal sources. The results of the regression analysis of this new model, carried out in the same fashion as described previously, are summarized in Table 4.

These results show that X_{1t} alone (model 1) cannot explain the changes since its corresponding Q_{96} due to SSE is above the $\chi^2(0.95;96) = 119.9$ bound. However, after initial inclusion of X_{2t}, model 3 has a large contribution from X_{1t}. As the value of Q_{96} due to SSE for model 3 (equation 22) is smaller than that corresponding to the previously selected model (equation 21), this model better explains the forecast residual variance, and also provides a reasonable physical explanation to the particulate concentration changes. The removal of particles due to the ban is estimated by (23)

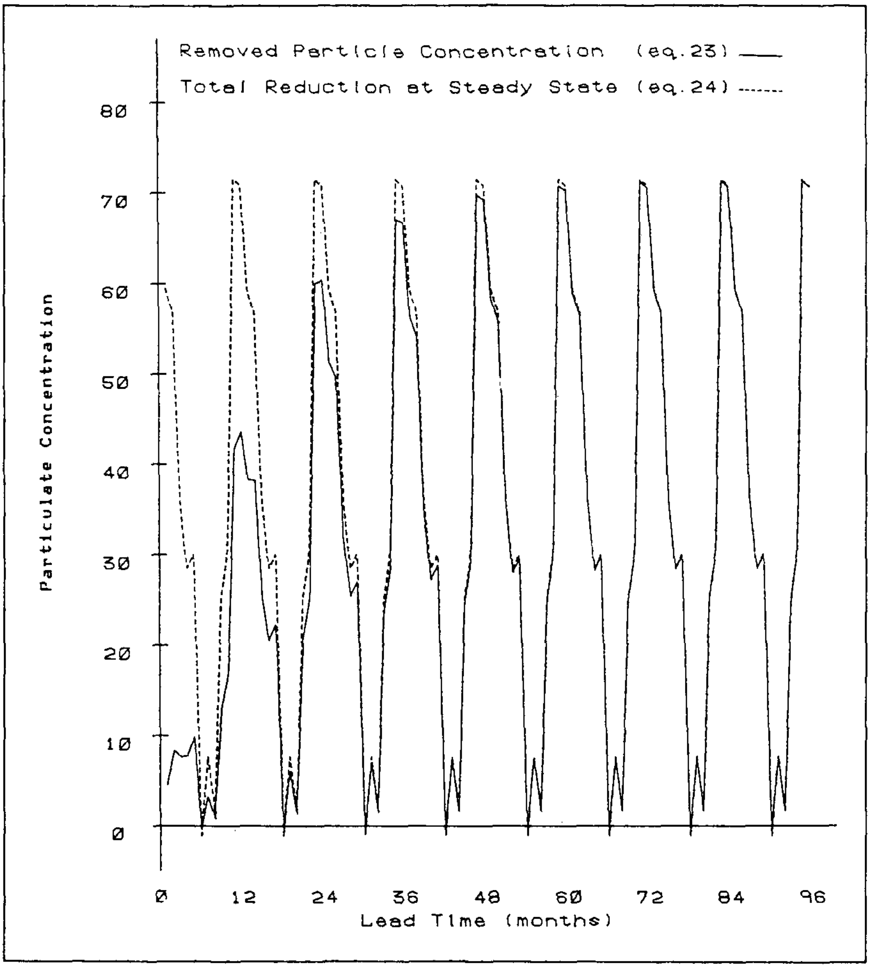


Figure 10. Particulate concentration due to high sulphur coal.

$$r_t = \beta_1 x_{1t} + \beta_2 x_{2t} . \tag{23}$$

The series w_t given in (24) represents the total monthly concentration of particulates due to burning of high sulphur coal in the period prior to the ban, so that the reduction in particles r_t brought about by the ban approaches steady state value w_t asymptotically.

$$w_t = \beta_1 s_1 + \beta_2 . \tag{24}$$

The time series of monthly reductions in particulate levels r_t is plotted together with the steady state reduction w_t in Figure 10.

VI. SUMMARY AND CONCLUSIONS

Changes in the monthly average air suspended particulate concentrations in the city of Chicago after the introduction of a law prohibiting the use of high sulphur coal have been analyzed. These changes have been shown to be statistically significant by means of a *portmanteau* χ^2 statistic based on the one-month ahead forecast errors. The changes in particulate concentrations are modelled by regression analysis using weight series corresponding to changes in parameters and time series of indicator variables.

Several alternative models of this kind have been analyzed. A model in which the change in particulate concentrations is composed of a change in the slope of the trend line and a change in the seasonal character of the series is first selected on the basis of its contribution to the χ^2 statistic. Then, an improved model that explains the changes in particulate concentration as a result of gradual reduction in the number of furnaces using high sulphur coal is constructed and analyzed. The results of the study permit the following conclusions:

1. The utility of the method of decomposition of the one-step ahead forecast errors to investigate the sources of changes in particulate concentration time series is clearly demonstrated in this study. The method is recommended for use in the study of the sources of changes in time series and in the development and validation of alternative models for explaining such changes.
2. The law that prohibits burning coal of high sulphur content has produced changes in the particulate concentrations in Chicago due to a gradual reduction in the emissions of particulates by furnaces using such coal. This reduction follows a pattern which approaches steady state after a lag of approximately five years. Among others, a model based on the first order reaction law adequately represents such a gradually changing pattern. This model could be used to investigate other environmental changes similar to that brought about by the law banning the burning of high sulphur coal which has been analyzed in this study.

APPENDIX A

A Taylor's expansion of the operator $\pi'(B)$ from (10) yields

$$\pi'(B) = \pi(B) + [\sum_{k=1}^{n} \Delta\gamma_k \frac{\partial}{\partial\gamma_k}]\,\pi(B) + \frac{1}{2!}[\sum_{k=1}^{n} \Delta\gamma_k \frac{\partial}{\partial\gamma_k}]^2\,\pi(B) + \ldots \quad (A.1)$$

The second and higher order terms are then dropped to obtain (A.2):

$$\pi'(B) = \pi(B) + \sum_{k=1}^{n} \Delta\gamma_k \frac{\partial\pi(B)}{\partial\gamma_k}. \quad (A.2)$$

Substitution of (11) and (A.2) in (10) gives

$$[\pi(B) + \sum_{k=1}^{n} \Delta\gamma_k \frac{\partial \pi(B)}{\partial \gamma_k}] [z_t - \sum_{j=1}^{m} \beta_j x_{jt}] = a_t . \quad (A.3)$$

After expanding, dropping the second order terms and rearranging, equation (A.4) is obtained.

$$\pi(B)z_t = \pi(B) \sum_{j=1}^{m} \beta_j x_{jt} - \sum_{k=1}^{n} \Delta\gamma_k \frac{\partial \pi(B)}{\partial \gamma_k} z_t + a_t . \quad (A.4)$$

Substituting a_t' for $\pi(B)z_t$ according to (6), (A.4) reduces to equation (A.5):

$$a_t' = \sum_{j=1}^{m} \beta_j \pi(B) x_{jt} - \sum_{k=1}^{n} \Delta\gamma_k \frac{\partial a_t'}{\partial \gamma_k} + a_t . \quad (A.5)$$

Equating the coefficients of β_j and $\Delta\gamma_k$ in (A.5) and (12), the expressions for X_{jt} and $W_{\gamma_k t}$ are obtained as equations (13) and (14), respectively.

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