Statistical Time Series Analysis of Emission and Deposition of SO2 and NOx in Northeastern North America

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1997

For Presentation at the Air & Waste Management Association's 90th Annual Meeting & Exhibition, June 8-13, 1997, Toronto, Ontario, Canada

97-A1130

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Abstract

Trend analysis and forecasting of time series data on air-pollutants is important to design effective measures to minimize damages to ecosystems and human health. In this study, autoregressive, moving average, autoregressive-moving average and autoregressive integrated moving average processes of different order were implemented to examine patterns of depositions and emissions. Analysis was undertaken to examine stationarity of the series or to design a method to create stationary series. The model that satisfied selected statistical criteria was chosen to make forecasts. Forecasts of depositions were compared with critical loads by watersheds.

The findings of this study indicated that both wet depositions and emissions of SO_2 and NO_x data exhibited non-stationarity. After removing non-stationarity, suitable time-series model was selected for short-run forecasting (1994 to 2005). The resulting depositions and emissions data were examined with respect to their long-run movement and critical deposition loadings. The analysis showed that excess wet depositions of SO_2 and NO_3 would be major problems at least for ten years. Most of these problems are observed in Atlantic Canada and few watersheds in Quebec and Ontario. Although emissions of SO_2 have declined, emissions of NOx remained unchanged or increased compared to the 1980 level. Considering the fact that these pollutants contribute to acidification, eutrophication and formation of secondary particulates that are hazardous to human health, it is necessary to find ways of further reducing emissions and depositions of these pollutants. While substantial progress has been made with

respect to reduction of SO_2 emissions (especially in Canada), the analysis presented in this study indicated that there must be substantially more reductions to ensure the protection of sensitive ecosystems. Thus, evidences similar to those presented in this study should be gathered to initiate negotiations for reductions beyond the 2005 or 2010 commitments.

Introduction

Environmental decision-makers and scientists realized that analysis of single pollutant assuming constancy to a host of other factors is not going to produce strategies that will improve the environment. The shift from single to multi-pollutant approach as well as the need to ensure sustainability also implies that socioeconomic considerations should be used to examine the feasibility of management strategies. All these, however, require large scale analysis. Often decision makers want important pieces of information on time. One such piece of information is analysis of by how much deposition or concentration are exceeding levels that would minimize effects to ecosystems and human health. The later can be obtained through statistical analysis of spatial and time-series information.

Once emission left the stack, meteorological factors (e.g., temperature, wind, etc.) are the primary determinants of dispersion and transport of pollutants. It is argued that these meteorological factors are uncertain and there is a high degree of randomness observed. The distribution mechanism underlying the statistical models also assumes randomness defined by specific parameters. Moreover, estimates and forecasts based on more than immediate distant time scale can very well reflect mean distribution of pollutants. Therefore, it may be possible to obtain results from time series analysis that may closely approximate those obtained from large-scale physical models.

Time series is a set of observation obtained by measuring a single variable regularly over a period of time. Using time series analysis, it is possible to discover systematic patterns in the series so that mathematical model can be built to explain the past behaviour of the series. Moreover, based on past behaviour, it is possible to forecast future values of the series.

A wide variety of questions are asked with respect to environmental variables. These questions range from simple average or mean value of a pollutant to quick short-run forecasts of depositions, concentrations, or exposure of resources to important pollutants. Time series models produce reliable data in short period of time. In situations where immediate decisions have to be made or important information is required for various reasons, analytical tools such as time-series models can be useful.

Analysis of the possible impacts of pollutants on forests, agriculture, water quality, fish population and human health can easily be ascertained though examination of exposure of certain geographic region to these pollutants. More importantly, the extent of resources at risk in the future can be derived from time series analysis. Wider application of time-series models to provide evidence on probable impact of pollutants on various elements of the ecosystem and human health can be ascertained at minimum cost, time and degree of complexity.

The problem

Environmental management principles are changing from mitigating impacts to precautionary or anticipate and act principle. This would require apriori analysis of environmental information so that decision makers would identify and implement informed and sound strategies. One such area of greater concern is related to analysis of resources or “stock” at risk. This includes analysis of deposition, concentration or emissions per unit of resources, including humans.

The practices of examining time-series environmental data often concentrate on moving average processes. While series may follow moving average of some order, there could also be other processes that may mimic the processes that generate the actual data and be used for forecasting. Techniques that are superior in their ability to estimate and forecast actual observations will also improve the uncertainty of environmental decision making.

In order to obtain reliable forecasts of emissions and depositions of pollutants, and to take appropriate measures before irreversible damages occur, it is essential to identify a model that mimics the dynamics of pollutants over time. This is very important in situations where physical models such as those involving diffusion equations to predict the spatial and temporal distribution of pollutants are not available in short period of time. On the other hand, nonphysical, or time-series models can be useful to provide information quite quickly.

The purpose of this study is to examine estimates and forecasts from three models moving averages, autoregressive, autoregressive moving average and autoregressive integrated moving averages processes. The performances of these techniques will be calibrated with respect to selected criteria, of which a method will be recommended for use in estimation and forecasting of these kinds of time-referenced data. Furthermore, actual and forecasted wet depositions will be compared with critical deposition loadings.[1]

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[1] Critical deposition loadings is define as: "The highest deposition of acidifying compounds that will not cause chemical changes leading to long term harmful effects on ecosystem structure and function" (Environment Canada, 1990). At the moment, critical loads are developed only for aquatic ecosystems.

Methodology

The techniques described below are well known in the field of econometrics time-series analysis. However, they are not widely used in the estimation and forecasting of time-series environmental variables. In this study, moving averages(MA), autoregressive(AR), autoregressive moving averages (ARMA), and autoregressive integrated moving averages(ARIMA) models are examined. The choice of a model to forecast a series is based on selected measures of fit.

In time series primary economic data (such as investment) or environmental data (such as emissions), the forces that generate the series may keep them together so that they would not drift apart. If a series is drifting apart, stationarity can be achieved of the series using various methods. It is essential to establish stationarity if the purpose is to examine true trends in the series and if there is a need to undertake forecasting.

In forecasting future values of a variable, time-series analysis relates the current values with past values, and current and past random disturbances. The unique feature of time-series analysis is that it doesn't begin with any conceptual framework provided say by economic theory. Instead, emphasis is placed on making use of information in the past values of a variable to forecast its future value. In the pages to follow, the methods of time-series analysis examined in this study will be described.

Time Series Models

Autoregressive (AR) Processes

Time series models assume that the future values of a variable depend on its past values plus random disturbances. An autoregressive model is based on the principle that past values, and past and current disturbances determine current values of a variable. Let an autoregressive process of order p be represented by, AR(p), then the equation is given by:

$$Y_t=\phi_1 Y_{t-1} + \phi_2 Y_{t-2} + \dots\dots\dots\dots + \phi_q Y_{t-p} + \delta + \varepsilon_t \qquad (1)$$

Where δ is an intercept parameter that relates to the mean of Y_t , ϕ_i's are unknown autoregressive parameters, ε_t is uncorrelated random error with zero mean and constant variance of σ^2_ε .

One of the problems in constructing the AR models is identifying the order of the underlying process. To identify the order of an AR process partial autocorrelation function is utilized. The sample and partial autocorrelation functions can be represented by Yule-walker equations that relate correlations in time t to past correlations. The Yule-Walker equations or autocorrelation functions are given:

$$\rho_1 = \phi_1 + \phi_2 \rho_1 + \dots\dots\dots \dots \dots\dots + \phi_p \rho_{p-1}$$

$$\begin{array}{ll} . & . \\ . & . \\ . & . \end{array}$$

$$\rho_p = \phi_1 \rho_{p-1} + \dots\dots\dots \dots\dots \dots \dots + \phi_p \qquad (2)$$

Solving the Yule-Walker equations for p will give us values of $\phi_1 \dots \phi_p$.

Solving equation 2 also requires knowledge of p. The partial autocorrelation function(PAF) could be derived by solving equation 2 for successive values of p. The PAF enables us to determine the order of the AR process. For example, if the order of a process is k then the PAF value should be close to zero for lags greater than k.

Moving Average (MA) Processes

Let's assume that change in the current values of a variable from year to year behave as a series of uncorrelated random variables with zero mean and constant variance. Let the series be Yt. Then,

$$Y_t = y_t - y_{t-1} = \varepsilon_t \text{ for } t=1....T \qquad (3)$$

Where ε_t is a random component.

The random component reflects new or unexpected issues, such as new information, unanticipated regulation affecting economic activity, unexpected wide spread use of new technology, etc. However, the full impact of any unexpected event may not be completely absorbed by current values of the variable. Thus, next year the value of the variable may be:

$$Y_{t+1} = \varepsilon_{t+1} + \theta\, \varepsilon_t \qquad (4)$$

Where ε_{t+1} is the effect of new information in year t+1 and $\theta\, \varepsilon_t$ reflect the impact from year t .

The representation given by equation (4) is a moving average process where the value of a variable in year t+1 is a weighted average of current and a past random variable.

In moving average process of order q, each observation Y_t, is generated by a weighted average of random disturbances going back q periods. Let's denote moving average process of order q by MA(q) and the equation becomes:

$$Y_t=\mu+\varepsilon_t -\theta_1\varepsilon_{t-1} - \theta_2\varepsilon_{t-2} -\theta_3\varepsilon_{t-3} \text{ ....} - \theta_q\varepsilon_{t-q} \qquad (5)$$

Where the parameters θ_1 to θ_q may be positive or negative. The disturbance terms are assumed to be independently and identically distributed across time ($\varepsilon \sim IID(0, \sigma_\varepsilon{}^2)$).

The order of the MA series can be identified using the autocorrelation function which enables us to determine at which lag the autocorrelation no longer differs from zero. For a moving average process of order q, the sample autocorrelation function should be close to zero for lags greater than q. The sample autocorrelation function is given by:

$$r_k= \sum_{t=1}^{t-k}(Y_t-Y^*)(Y_{t+k} -Y^*)/\sum_{t=1}^{t}(Y_t-Y^*)^2 \qquad (6)$$

Where Y* is the mean of the sample series.

Autoregressive Moving Average (ARMA) Processes

An ARMA model exhibits both MA and AR processes. A process with MA(q) and AR(p) denoted as ARMA(p,q) is given by:

$$Y_t=\phi_1Y_{t-1} + \phi_2Y_{t-2} + \text{......} + \phi_qY_{t-p} + \delta + \varepsilon_t -\theta_1\varepsilon_{t-1} - \theta_2\varepsilon_{t-2} -\theta_3\varepsilon_{t-3} \text{ ....} - \theta_q\varepsilon_{t-q} \qquad (7)$$

Integrated Series

The above procedures regarding estimation using AR, MA and ARMA processes assume that the series is stationary. The only concern is to identify the order of the process for the purpose of forecasting future values of a variable. However, non-stationary series are ubiquitous. In many cases a series could exhibit a monotonically upward or downward movement. Thus, the assumption of a constant mean upon which the above time-series models were based will be violated. The variance of a series may also become non-constant or infinite. These kinds of non-stationary series could be transformed such as by differencing so that the series could be made stationary. The number of times a series is differenced to be stationary indicates the order of integration. If a series Y_t is stationary after differencing d times, then it is said to be integrated of order d(see Engle and Granger, 1987).

Y_t is said to be nonstationary of order d if :

$$W_t=\Delta^d Y_t \qquad (8)$$

Where W_t is stationary series and Δ denotes differencing.

Summing the series W_t d times will give:

$$Y_t= \Sigma^d W_t \qquad (9)$$

The values of a variable Y_t can be represented as:

$$Y_t = Y_0 + W_1 +W_2 +W_3 \,..... \, W_t \qquad (10)$$

where Y_0 is the original undifferenced series, $W_t=\Delta Y_t$

Autoregressive Integrated Moving Average (ARIMA) Processes

ARIMA is a model that incorporates both autoregressive and moving average processes.

If $W_t = \Delta^d Y_t$, and W_t is an ARMA(p,q) process, then Y_t is an integrated autoregressive moving average process of order (p,d,q). ARIMA(p,d,q) can be written, using a backward shift operator, as:

$$\phi(B)\Delta^d Y_t =\delta+ \theta(B)\varepsilon_t \qquad (11)$$

With $\phi(B)=1-\phi_1 B-\phi_2 B^2$ -..............- $\phi_p B^p$

$\theta(B)=1-\theta_1 B-\theta_2 B^2$ -..............- $\theta_p B^p$ (12)

$\phi(B)$ is called the autoregressive operator and $\theta(B)$ the moving average operator.

There are several estimating and forecasting techniques of time-series variables. Many of these techniques are fitted to a data on the assumption that the model is an adequate approximation to the true generating mechanisms and then forecasts are made using the model. Among most models used to forecast time series data, the ARIMA has been found to be superior (see Granger and Engle, 1987)

Testing for Stationarity (Unit-Root)

Estimation of AR, MA and ARMA processes apply only to stationary time series. If the data is non-stationary, it implies that it contains an integrated component and that it should be differenced either before or during estimation process.

A series is called weakly stationary if it has finite mean, a finite variance and covariances, all of which are independent of time. Let's consider an AR(1) process:

$$\Delta Y_t = \mu + \rho Y_{t-1} + \varepsilon_t \qquad (13)$$

where μ and ρ are parameters and the ε_t 's are assumed to be independently and identically distributed with zero mean and equal variance. If $|\rho|$ is between -1 and 1, the series is stationary. If $|\rho| =1$ the equation defines a random walk with drift and Y is then non-stationary. The variance of a series with a unit root becomes infinite. If $|\rho| >1$ then the series is explosive. Thus, the null hypothesis for testing non-stationarity is that $|\rho| =1$. The null hypothesis is,

H_0: $\rho=1$

The test of this hypothesis is called a unit root test. If a series is represented by:

$$\Delta Y_t = \mu + \Upsilon Y_{t-1} + \varepsilon_t \qquad (14)$$

where $\Upsilon=\rho-1$. Thus, the null hypothesis is H_0: $\Upsilon=0$. Rejection of the hypothesis implies stationarity.

Measures of Model Fitness

Forecasting time series variables requires that the model selected has adequately fit the data. To ensure accuracy of the forecast, the models have to be screened using various measures of fitness. In the present study, Akaike Information Criterion (AIC) and Schwarz Bayesian Criterion (SBC) will be used in addition to standard errors. A model with minimum values of these measures is hypothesized to be the best candidate for use in forecasting. These measures (AIC and SBC) are also helpful to decide the order of the model. AIC and SBC take into account both how well the model fits the observed series, and the number of parameters used in the fit.

Sources of Data

SO_2 and NO_3 wet deposition, as well as emission data were examined in the present study. The data on wet deposition covering the period 1980 to 1993 was obtained from the Atmospheric Environment Research Services of Environment Canada. Emission data was gathered from OECD (1995) and Environment Canada (1996). Critical Deposition loadings for SO_2 are obtained from Environment Canada (1993). These CLs are for wet depositions. However, review of publications (see Jefferies, 1995) indicate that nitrogen CLs may be less than that of SO_2. Therefore, CLs for SO_2 were used in the analysis of exceedances for wet NO_3 depositions. Thus, the results of exceedances analysis for NO_3 should be interepreted with caution.

The deposition data was overlayed over watershed aggregates located in eastern Canada using GIS software (see Figure 1). These watersheds are numbered 1 through 22. In the present study, watershed # 6 was excluded because of insufficient information. Average values of wet deposition were calculated using the GIS software. This data was used to examine trends and make forecasts of depositions.

Results of the Analysis

Tests for Stationarity (Unit Root)

There are two commonly used tests of unit-root: Augmented Dickey-Fuller (ADF) and Phillips-Perron tests. In the present study the ADF tests were used. Unit root tests using the Augmented Dickey-Fuller test for wet NO_3 depositions indicated that the series for all but Watershed14, were found to be non-stationary. The results for wet SO_2 depositions indicate that all series exhibited non-stationarity. After differencing, all depositions data for all watersheds exhibited stationarity. Similarly, the emissions data showed non-stationarity. Therefore, the series were differenced to establish stationarity. Once, stationarity was established, AR, MA and ARMA models were estimated. The model with the smallest value of standard error, AIC and SBC was selected to make forecasts for the period 1994 to 2005. The degree of differencing and estimation method is summarized in Table 1.

Results of Time Series Analysis

Emissions of NOx showed a slight increase or remained unchanged until the year 2005 for both Canada and US compared to the 1980 level (see Table 2). However, emission of SO_2 showed a substantial reduction for Canada and almost 30% reduction for the USA in 2010 compared to the 1980 level. Forecast of emissions by Environment Canada, taking into account economic growth factors, indicated an increase of NOx emissions by 8% but a decline of SO_2 emissions by 38%. The time series model used in the present study underestimated the Environment Canada's projection of NO_X emissions by less than 5%. However, due to drastic reductions of SO_2 beginning in the late 80's till 1994, the time series model underestimated Environment Canada's projection of SO_2 by about 30% (see Table 2). Projection of the US emission also indicated that NOx emissions will decline by about 12% while SO_2 emissions will decline by about 37% compared to the 1980 level (Canada-US Air Quality Agreement, 1994). The forecast from the present study overestimated NOx and SO_2 emissions by about 10%. The difference between the values of forecast from the present study and those from official documents is that the latter are based on large scale study taking into account exogenous factors in making the forecast. The time-series models, however, rely only on past values of the variable. Based on these differences and the need to have environmental information quickly and cheaply, the over or underestimation could be considered relatively small. Future analysis of data on pollutants will consider other more sophisticated tools so that radical changes in the pattern of the observed series could be better accounted for and makes forecasts relatively superior by incorporating important exogenous factors such as location, regulations, technologies, etc.

The conclusion is that while some degree of success has been achieved with respect to SO_2 emissions, a lot more has to be done with respect to emission of NOx. Considering the fact that either of these pollutants contributes to acid rain, eutrophication and formation of particulates, it is necessary to find ways of further reduction emissions of NOx and SO_2.

Table 1. Summary of Degree of Differencing and Estimation Methods For Wet NO_3 and SO_2 Depositions[2]

Watershed Number	Estimation Method for NO_3	Estimation Method for SO_2
1	ARIMA (0,1,1)	ARIMA (0,2,1)
2	ARIMA (1,1,1)	ARIMA (1,2,1)
3	ARIMA (0,2,1)	ARIMA (1,2,1)
4	ARIMA (1,1,1)	ARIMA (0,1,1)
5	ARIMA (1,2,1)	ARIMA (1,2,0)
7	ARIMA (1,2,1)	ARIMA (0,2,1)
8	ARIMA (1,1,1)	ARIMA (1,1,0)
9	ARIMA (0,2,1)	ARIMA (1,2,1)
10	ARIMA (0,1,1)	ARIMA (1,1,1)
11	ARIMA (0,1,1)	ARIMA (1,2,1)
12	ARIMA (1,2,1)	ARIMA (1,2,1)
13	ARIMA (1,1,0)	ARIMA (1,1,1)
14	ARIMA (1,0,1)	ARIMA (1,2,1)
15	ARIMA (1,1,0)	ARIMA (1,2,1)
16	ARIMA (0,2,1)	ARIMA (1,2,1)
17	ARIMA (0,1,1)	ARIMA (1,1,1)
18	ARIMA (1,2,1)	ARIMA (1,2,1)
19	ARIMA (1,2,1)	ARIMA (1,2,1)
20	ARIMA (1,2,0)	ARIMA (1,2,1)
21	ARIMA (0,1,1)	ARIMA (1,1,1)
22	ARIMA (1,2,0)	ARIMA (1,2,1)
Country	**Emissions of NOx**	**Emissions of SO_2**
Canada	ARIMA(1,2,1)	ARIMA(1,1,1)
USA	ARIMA(0,2,1)	ARIMA(1,2,1)

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[2] The numbers separated by a comma in the brackets in Columns 2 and 3 following the abbreviation ARIMA indicate the order of Autoregressive process, Differencing and Moving average process respectively.

Table 2. Actual (1980-1993) and Forecasted (1994-2005)Emissions of Nitrogen and Sulphur Oxides in Canada and the USA, in Kilotonnes

	Based on the present study				Forecast of Emissions[3]			
Year	CANNOx	$CANSO_2$	$USNO_x$	$USSO_2$	$CanNO_x$	$USNO_x$	Can SO_2	$USSO_2$
1980	1959.000	4643.000	21469.000	23779.000				
1981	1907.000	4291.000	21315.000	22512.000				
1982	1897.000	3612.000	20571.000	21212.000				
1983	1884.000	3625.000	19967.000	20619.000				
1984	1871.000	3955.000	20526.000	21467.000				
1985	1984.000	3692.000	20338.000	21219.000				
1986	1934.000	3627.000	20214.000	20391.000				
1987	2037.000	3762.000	20694.000	20519.000				
1988	2117.000	3838.000	21440.000	20948.000				
1989	2120.000	3695.000	21299.000	21043.000				
1990	1999.000	3323.000	21373.000	20701.000				
1991	1976.000	3306.000	21240.000	20660.000				
1992	1939.000	3030.000	21001.000	20622.000				
1993	1980.000	3035.000	21240.000	19518.000				
1994	1995.000	2668.000	21500.000	19200.000				
1995	1987.294	2490.140	21359.525	19999.396	1999		2805	
1996	1981.067	2368.646	21170.178	19432.107				
1997	1977.883	2209.330	21086.609	19151.946				
1998	1976.255	2075.393	21049.725	18811.463				
1999	1975.422	1924.427	21033.447	18483.652				
2000	1974.997	1784.886	21026.262	18153.179	2080	18500	2802	15700
2001	1974.779	1637.680	21023.091	17823.266				
2002	1974.668	1495.617	21021.692	17493.235				
2003	1974.611	1350.103	21021.074	17163.228				
2004	1974.581	1206.905	21020.802	16833.217				
2005	1974.567	1062.152	21020.681	16503.206	2121	18700	2854	15000

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[3] Data for Columns 2 to 5 are obtained from OECD (1995), Environment Canada (1996) and Air Quality Committee (1996), while data for columns 6 to 9 are obtained from Air Quality Committee (1996) and Environment Canada (1996).

Comparison of Actual Versus Critical Deposition loadings by Watersheds

Comparison of actual and forecasted depositions of NO_3 and SO_2 indicated substantial variations. In general, there is a declining trend of the area covered by depositions of more than 20kg/ha/yr. However, the visual presentations show that depositions of SO_2 and NO_3 could be environmental problems at least for the next ten years (see Figures 2,3,4,5,6 and 7).

Wet Depositions of NO_3 and Critical Loads

Comparison of wet depositions of NO_3 and critical deposition loadings are presented in Table 3. The results indicate that several watersheds enjoyed depositions less than critical loads. However, watersheds 1, 2,3,4,7,8, (in the Atlantic regions), watersheds 12 and 14 (in Quebec), and watersheds 16,17 and 19 (in Ontario) will have excess wet depositions until the year 2005. Moreover, some watersheds such as watershed #5 tend to show a tendency toward increased depositions making the gap between actual and critical deposition loadings narrower. In fact, projections to the year 2010 indicated that this watershed will experience excess depositions. These watersheds cover substantial area of Eastern Canada with a population of at least three million. Consequently, large amounts of resources and million of people may be exposed to the impact of excess depositions of NO_3. If depositions, measured in static terms, are not reduced at a faster rate, the damages to ecosystems and human health could be very large. However, background depositions of SO_2 in Eastern Canada are between 5 to 6 kg/ha/yr (Environment Canada, 1993). Thus, depositions that are less than CLs by more than 5kg/ha/yr should be a warning for another kind of environmental problem.

Emission forecasts seem to indicate a general declining trend. However, excess depositions in some watersheds exhibit increasing depositions and possibly acidity until 2005. This observation may be due to the fact that the cumulative nature of depositions whereby a small amount of increases over previous no-neutralized deposition may show increases over time.

Wet Depositions of SO_2 and Critical Loads

Most watersheds seem receive depositions less than critical loads (Table 4). However, watersheds 4,5,7 and 8 (Atlantic regions) and watersheds 12 and 14 (Quebec) observed excess depositions for the duration of the forecast.

Table 3. Exceedenaces of Actual Wet over Critical Deposition Loadings for NO_3 (kg/ha/yr)

Year/ Watershed	1	2	3	4	5	7	8	9	10	11	12	13
1980	2.118	-1.177	2.245	-1.898	-4.733	-4.460	-4.342	-5.604	-4.133	-5.047	-0.987	0.853
1981	2.386	-1.371	0.454	-0.844	-5.040	-4.454	-4.371	-5.926	-4.742	-5.604	-3.531	-0.599
1982	-0.066	-1.741	-0.422	-1.443	-4.004	-3.637	-3.617	-4.603	-3.627	-4.119	2.188	2.623
1983	-2.020	-2.070	-0.001	-1.780	-4.521	-3.098	-2.977	-3.266	-2.172	-2.665	0.276	-0.670
1984	0.126	-0.542	0.419	0.415	-4.273	-2.223	-1.872	-3.319	-3.145	-4.481	1.670	-0.222
1985	3.165	1.841	0.508	-1.409	-4.804	-3.273	-3.343	-4.998	-4.068	-5.274	1.764	-0.688
1986	3.106	1.266	0.284	-1.262	-4.498	-3.083	-3.215	-5.020	-4.849	-5.902	0.411	-1.990
1987	0.837	-0.819	-2.029	-3.470	-3.965	-3.562	-3.528	-4.641	-5.207	-5.946	-1.922	-2.082
1988	1.928	0.713	2.413	0.448	-4.502	-1.359	-1.718	-5.027	-3.872	-5.226	3.024	0.726
1989	6.726	4.108	2.829	2.707	0.143	-0.418	0.244	-2.147	-3.484	-5.158	1.829	0.983
1990	3.732	1.517	3.651	3.335	-0.868	1.491	2.438	-1.193	-2.390	-4.483	5.336	2.085
1991	1.980	1.704	1.031	2.439	-2.549	-0.544	-0.154	-3.275	-3.915	-5.324	0.784	-1.780
1992	1.580	0.740	0.359	0.769	-2.501	-0.721	-1.105	-3.919	-3.689	-4.857	-0.613	-1.387
1993	1.500	0.245	1.411	1.966	-1.745	-0.208	-0.299	-4.132	-3.371	-5.044	1.818	0.331
1994	2.074	0.354	1.103	2.263	-2.659	0.119	0.012	-3.815	-3.670	-5.133	2.034	-0.108
1995	1.931	0.463	1.018	2.560	-2.842	0.446	0.323	-4.111	-3.756	-4.944	2.250	-0.125
1996	1.931	0.573	0.994	2.858	-2.744	0.773	0.634	-4.111	-3.756	-4.944	2.465	-0.126
1997	1.931	0.682	0.988	3.155	-2.540	1.100	0.945	-4.111	-3.756	-4.944	2.681	-0.126
1998	1.931	0.791	0.986	3.452	-2.294	1.427	1.256	-4.111	-3.756	-4.944	2.897	-0.126
1999	1.931	0.901	0.986	3.749	-2.032	1.755	1.567	-4.111	-3.756	-4.944	3.113	-0.126
2000	1.931	1.010	0.985	4.047	-1.765	2.082	1.878	-4.111	-3.756	-4.944	3.328	-0.126
2001	1.931	1.119	0.985	4.344	-1.495	2.409	2.189	-4.111	-3.756	-4.944	3.544	-0.126
2002	1.931	1.229	0.985	4.641	-1.224	2.736	2.500	-4.111	-3.756	-4.944	3.760	-0.126
2003	1.931	1.338	0.985	4.938	-0.953	3.063	2.811	-4.111	-3.756	-4.944	3.976	-0.126
2004	1.931	1.448	0.985	5.236	-0.681	3.390	3.122	-4.111	-3.756	-4.944	4.192	-0.126
2005	1.931	1.557	0.985	5.533	-0.410	3.717	3.433	-4.111	-3.756	-4.944	4.407	-0.126

Table 3. (Continued)

Year/ Watershed	14	15	16	17	18	19	20	21	22
1980	5.217	-1.605	7.931	3.964	3.481	8.295	-13.63	-4.217	-8.005
1981	2.056	-5.656	5.204	2.831	2.897	3.558	-14.60	-4.184	-10.06
1982	7.758	-3.688	2.053	2.700	1.416	4.456	-13.14	-3.029	-7.380
1983	5.456	-3.965	2.360	1.117	0.390	6.080	-13.68	-3.076	-9.400
1984	4.401	-2.918	4.670	3.700	3.112	6.928	-13.67	-2.707	-9.800
1985	4.971	-3.528	5.359	5.244	5.278	6.267	-13.81	-1.950	-8.897
1986	3.000	-4.651	4.643	1.297	3.228	4.941	-14.28	-2.915	-10.60
1987	2.915	-5.674	4.636	1.651	2.132	4.599	-13.46	-2.806	-9.517
1988	7.682	1.197	6.236	6.902	2.115	12.191	-10.89	-0.872	-8.110
1989	6.949	-1.835	5.655	3.226	1.634	6.856	-13.48	-2.667	-10.00
1990	6.426	-2.164	5.060	4.751	1.730	8.606	-12.82	-2.609	-7.473
1991	2.833	-5.043	4.803	2.167	-0.273	6.053	-13.14	-1.943	-9.269
1992	3.616	-4.481	5.525	2.349	2.137	7.653	-12.97	-2.606	-9.987
1993	5.186	-3.776	6.631	5.084	-0.023	7.245	-13.11	-3.653	-7.805
1994	5.251	-3.943	6.106	2.766	-0.292	7.164	-17.32	-3.235	-9.738
1995	5.104	-4.110	5.603	3.116	-0.562	7.083	-17.07	-2.746	-8.741
1996	5.026	-4.277	5.440	3.249	-0.831	7.003	-16.99	-2.860	-9.255
1997	4.985	-4.444	5.387	3.300	-1.101	6.922	-16.90	-2.833	-8.990
1998	4.963	-4.612	5.370	3.319	-1.370	6.841	-16.80	-2.840	-9.127
1999	4.951	-4.779	5.365	3.327	-1.640	6.760	-16.71	-2.838	-9.057
2000	4.945	-4.946	5.363	3.329	-1.909	6.680	-16.62	-2.838	-9.093
2001	4.942	-5.113	5.362	3.330	-2.179	6.599	-16.53	-2.838	-9.074
2002	4.940	-5.280	5.362	3.331	-2.448	6.518	-16.44	-2.838	-9.084
2003	4.939	-5.447	5.362	3.331	-2.718	6.437	-16.35	-2.838	-9.079
2004	4.939	-5.614	5.362	3.331	-2.987	6.357	-16.26	-2.838	-9.081
2005	4.939	-5.781	5.362	3.331	-3.257	6.276	-16.16	-2.838	-9.080

Table 4. Exceedances of Actual Wet over Critical Deposition Loadings for SO_2 (kg/ha/yr)

Year/ watershed	1	2	3	4	5	7	8	9	10	11	12	13
1980	12.182	4.806	12.301	6.468	3.385	4.111	4.358	3.280	5.244	3.815	9.381	10.170
1981	12.934	4.909	10.646	8.197	1.208	2.927	3.193	1.428	4.051	2.551	7.196	9.056
1982	7.652	3.376	6.191	4.312	0.652	0.487	0.655	-1.655	-0.262	-2.672	9.521	10.426
1983	3.519	2.790	4.540	1.840	0.592	2.838	2.498	0.712	0.871	0.181	6.054	3.179
1984	6.667	4.985	7.379	8.694	3.195	5.180	5.366	2.224	0.629	-1.483	8.134	4.081
1985	8.178	7.494	6.326	6.973	0.270	1.180	1.169	-2.266	-2.190	-4.353	6.185	2.765
1986	9.036	7.069	6.498	5.322	1.231	1.014	1.152	-1.747	-2.434	-4.717	7.524	1.840
1987	4.674	4.375	3.155	1.634	0.511	1.082	1.521	-0.417	-2.007	-3.909	3.039	3.330
1988	6.062	4.215	6.236	5.765	1.157	4.381	5.320	1.685	-1.035	-4.868	7.221	3.115
1989	10.004	5.652	5.548	7.161	3.550	2.635	3.046	-0.988	-1.668	-4.204	3.779	3.419
1990	8.587	3.787	8.374	8.874	2.846	5.022	5.880	0.662	0.334	-1.929	10.242	7.821
1991	6.611	5.532	5.388	10.646	1.697	3.742	3.926	-1.983	-2.234	-4.448	4.680	2.309
1992	3.842	3.130	4.374	7.292	1.705	3.645	2.977	-1.312	-0.304	-2.295	1.979	2.569
1993	3.445	1.827	4.270	5.750	2.253	2.960	2.687	-2.872	-2.717	-4.753	4.156	2.753
1994	2.773	-1.109	3.652	6.215	1.739	2.948	3.115	-0.525	-2.563	-5.413	6.470	2.182
1995	2.101	-2.837	3.034	6.315	1.734	2.948	3.126	-0.258	-3.376	-6.072	6.362	1.612
1996	1.429	-3.880	2.416	6.337	1.734	2.948	3.126	-0.228	-3.837	-6.731	6.362	1.041
1997	0.757	-4.534	1.798	6.341	1.734	2.948	3.126	-0.225	-4.426	-7.390	6.362	0.471
1998	0.085	-4.969	1.181	6.343	1.734	2.948	3.126	-0.224	-4.969	-8.049	6.362	-0.100
1999	-0.587	-5.278	0.563	6.343	1.734	2.948	3.126	-0.224	-5.528	-8.708	6.362	-0.670
2000	-1.259	-5.517	-0.055	6.343	1.734	2.948	3.126	-0.224	-6.081	-9.367	6.362	-1.241
2001	-1.931	-5.715	-0.673	6.343	1.734	2.948	3.126	-0.224	-6.637	-10.026	6.362	-1.811
2002	-2.603	-5.891	-1.291	6.343	1.734	2.948	3.126	-0.224	-7.192	-10.685	6.362	-2.382
2003	-3.275	-6.054	-1.909	6.343	1.734	2.948	3.126	-0.224	-7.747	-11.344	6.362	-2.952
2004	-3.947	-6.209	-2.526	6.343	1.734	2.948	3.126	-0.224	-8.302	-12.003	6.362	-3.523
2005	-4.619	-6.361	-3.144	6.343	1.734	2.948	3.126	-0.224	-8.856	-12.662	6.362	-4.093

Table 4. (Continued)

Year/ watershed	14	15	16	17	18	19	20	21	22
1980	13.931	6.108	16.610	10.725	12.808	14.226	-9.808	-1.490	-1.340
1981	12.213	6.112	16.217	13.936	17.077	14.822	-10.030	-1.625	-2.966
1982	14.711	2.499	10.549	10.253	12.717	11.596	-7.568	0.734	0.067
1983	13.198	4.257	9.788	9.185	10.334	13.734	-9.829	-2.197	-5.148
1984	9.940	4.085	11.446	11.015	12.318	15.364	-9.865	-1.429	-4.751
1985	10.923	1.382	9.006	10.078	13.643	12.369	-2.411	-0.808	-2.804
1986	9.578	1.961	12.236	5.877	10.879	10.224	-12.210	-2.734	-7.059
1987	10.651	0.135	9.942	3.898	8.905	9.296	-11.630	-3.359	-3.174
1988	11.797	5.623	9.943	8.397	7.553	14.761	-9.799	-0.867	-6.126
1989	7.437	-0.993	6.002	3.927	4.612	9.507	-12.040	-2.550	-6.821
1990	9.973	1.022	9.380	7.530	8.232	13.337	-11.710	-3.113	-2.015
1991	6.916	-0.542	8.570	5.099	3.751	12.395	-12.180	-1.850	-6.657
1992	6.526	-1.909	8.162	4.555	4.960	11.239	-11.470	-1.765	-6.766
1993	7.214	-1.603	8.125	5.133	1.389	10.843	-12.327	-3.597	-5.018
1994	9.423	-16.845	-13.187	0.853	-11.837	-7.363	-20.868	-10.189	-12.144
1995	10.391	-11.869	-18.768	0.432	-10.257	-5.123	-21.750	-8.908	-10.041
1996	10.391	-14.501	-20.540	-0.183	-11.749	-5.682	-22.043	-9.363	-11.229
1997	10.391	-14.271	-21.390	-0.788	-12.603	-5.858	-22.291	-9.435	-11.243
1998	10.391	-15.117	-22.017	-1.393	-13.589	-6.087	-22.535	-9.591	-11.676
1999	10.391	-15.559	-22.590	-1.999	-14.548	-6.308	-22.779	-9.729	-11.959
2000	10.391	-16.152	-23.149	-2.605	-15.513	-6.531	-23.023	-9.871	-12.296
2001	10.391	-16.689	-23.705	-3.210	-16.476	-6.753	-23.268	-10.013	-12.614
2002	10.391	-17.247	-24.261	-3.816	-17.440	-6.975	-23.512	-10.154	-12.938
2003	10.391	-17.797	-24.816	-4.422	-18.404	-7.198	-23.756	-10.295	-13.260
2004	10.391	-18.350	-25.371	-5.027	-19.368	-7.420	-24.000	-10.437	-13.583
2005	10.391	-18.901	-25.926	-5.633	-20.331	-7.642	-24.244	-10.578	-13.906

The results for wet depositions of NO_3 and SO_2 indicate that most regions in Atlantic Canada, some portion of Quebec and Ontario will suffer from the impacts of acidic deposition despite the efforts of the government and industry to reduce emissions in both the US and Canada. These findings imply that there has to be further negotiation on additional reduction plans so that watersheds that are experiencing severe acidic problems from could be protected from further damages. Moreover, other impacts such as eutrophication and particulate matter which are believed to be significantly influenced by emissions of NOx and SO_2 can be minimized.

Conclusions

Time series models such as AR, MA, ARMA and ARIMA were applied to NO_3 and SO_2 deposition, and NOx and SO_2 emissions data for the US and Canada. The results indicate that i) most series are not stationary and required differencing to establish stationarity, ii) after differencing each series corresponding to different watersheds or country was best approximated by either AR, MA or ARMA processes, iii) emissions of NOx tend to increase slightly or remained unchanged, iv) several watersheds in Atlantic Canada and few watersheds in Southeastern portions of Quebec and Ontario will experience excess acidic depositions between the period 1994 to 2005.

Despite the commitments of the Canadian and US governments, the impact of SO_2 and NOx emissions are anticipated to prevail for the next ten years. More importantly, the dynamic analysis or cumulative depositions, may show even larger excess depositions in many watersheds. Therefore, it is necessary to find ways of further reducing emissions of these pollutants to protect sensitive ecosystems.

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