

Tropospheric Ozone Trend in the Muda Irrigation Area, Kedah

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Abstract: *The ozone trend in the Muda Irrigation Area (MADA), Kedah, Malaysia, was assessed from the ozone data recorded in Sungai Petani. Three parameters were selected as the robust trend indicators in the study: the monthly mean, the monthly averaged daily 1-h maximum and the number of annual hours $> 120 \mu\text{g}/\text{m}^3$. As the ozone data displayed obvious seasonal variation, using deseasonalised monthly average parameters to estimate the ozone trends could smooth out the influence of seasonal fluctuations. In this study, we used the Box-Jenkins methodology to build the Auto-Regressive Integrated Moving Average (ARIMA) model for the monthly ozone data taken from an Automatic Air Quality Monitoring System in Sungai Petani station for the period between 1999 and 2007, with a total 108 readings. The parametric, seasonally adjusted ARIMA $(1, 0, 1) \times (2, 1, 2)^{12}$ model was successfully applied to predict the long-term trend of ozone concentration. The detection of a steady, statistically significant upward trend of ozone concentration in the area is a concern for human health and agricultural activities.*

Keywords: Tropospheric ozone, time series analysis, seasonal variation, MADA area

Abstrak: *Trend ozon troposfera di Kawasan Pengairan Muda (MADA), Kedah telah dinilai daripada data ozon yang telah direkodkan di Sungai Petani. Tiga parameter telah dipilih sebagai penunjuk tetap kajian ini: min bulanan, purata bulanan 1-j maksimum setiap hari dan bilangan jam tahunan $> 120 \mu\text{g}/\text{m}^3$. Data ozon menunjukkan kepelbagaian bermusim dengan jelas, manakala parameter purata bulanan tidak bermusim pula digunakan untuk menyingkirkan pengaruh perubahan musim. Dalam kajian ini, kami menggunakan metodologi Box dan Jenkins untuk membina model data bulanan ozon yang diperolehi dari Sistem Pemantauan Kualiti Udara Otomatik di stesen Sungai Petani dari 1999 hingga 2007 dengan jumlah 108 bacaan data. Model berparameter ARIMA terlaras secara bermusim $(1, 0, 1) \times (2, 1, 2)^{12}$ telah berjaya diaplikasi bagi meramal arah aliran jangka panjang kepekatan ozon. Pengesanan secara mantap tambahan berkesan arah aliran meningkat bagi kepekatan ozon di kawasan ini adalah membimbangkan.*

Kata kunci: Ozon troposfera, analisis siri masa, kepelbagaian bermusim, kawasan MADA

1. INTRODUCTION

It has long been recognised that pollutant gases cause significant impacts on crops. Studies have revealed that ground-level ozone is responsible for most of the crop-yield losses from air pollutants, while losses from other pollutants are minimal relative to ozone.¹⁻³ Exposure to tropospheric ozone creates adverse effects on human health, the ecosystem and crops. Several studies have shown that ozone reduces yields in crops such as tobacco, beans, spinach, watermelon, wheat and other cereals.^{4,5} In Spain, the effects of ozone levels on citrus crops have been assessed at the "La Plama de Castellon," and the results revealed estimated loss percentages in crop harvests of approximately 10% for lemon trees and 8% for orange trees.⁵ In the stratosphere, atomic oxygen is formed by the photo-dissociation of molecular oxygen, while the photolysis of nitrogen dioxide is the main source providing oxygen in the troposphere.⁶ In the troposphere, ground level ozone is a secondary pollutant that forms from photochemical reactions primarily between volatile organic compounds (VOCs) and nitrogen oxides (NOx).

Despite massive and costly control efforts, countries in Europe and North America are still experiencing severe ozone problems.⁷ People in Asia cannot escape from ozone pollution. Elevated ozone levels have been reported in some large Asian cities.⁸ Nevertheless, the long-term ozone trend, especially in Malaysia, is relatively less researched. This omission is serious because of the greater importance of this issue in developing countries due to the increasing demand for higher crop production in the face of growing populations and the rapid deterioration of ambient air quality associated with industrialisation and urbanisation as well as land constraints.⁹ Moreover, Malaysia is located at the equatorial region and is at greater risk because its climate is characterised by high levels of solar radiation, which can promote the formation of photochemical pollutants, such as ozone (O₃).¹⁰

This study aims to extend ozone trend analysis to provide both qualitative and quantitative information about ozone concentrations in the Muda Irrigation Scheme Area (MADA) and to predict future concentrations of this pollutant. MADA is considered the "rice bowl" of Malaysia as it contributes to 40% of the total rice production in Malaysia.¹¹ Detection of the trend will enable decision makers to establish priorities in terms of air pollution management.

2. EXPERIMENTAL

2.1 Study Area

This study was conducted in Sungai Petani ($6^{\circ} 11.8' N$, $100^{\circ} 4.5' E$), a developing Malaysian town located in MADA, Kedah, northwest of Peninsular Malaysia (Figure 1). This area is where dominant sources of ozone precursors related to industrial activities and road traffic are found.

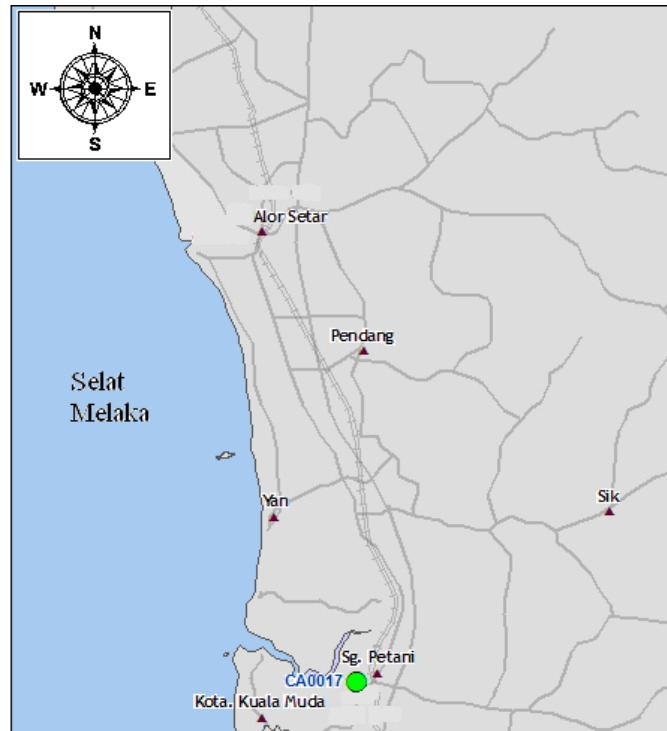


Figure 1: Location of the study area.

2.2 Data and Monitoring Network

The monitoring network was installed, operated and maintained by Alam Sekitar Malaysia Sdn. Bhd. (ASMA) through concessions by the Department of Environment Malaysia.¹² The tropospheric ozone concentration data were recorded using a system manufactured by Teledyne Technologies Incorporated (Model 400E), which is based on the Beer-Lambert law for measuring low ranges

of ozone in ambient air. In this instrument, a 254-nm UV light signal is passed through the sample cell, where it is absorbed in proportion to the amount of ozone present. In this study, the ozone trend was examined using ozone data consisting of 108 monthly observations from January 1999 to December 2007 acquired from the Air Quality Division of ASMA for Sekolah Menengah Kebangsaan Tun Ismail, Bakar Arang, Sungai Petani.

2.3 Time Series Analysis

Time series analysis was implemented using the StatGraphics statistical software package. A time series consists of a set of sequential numeric data taken at equally spaced intervals, usually over a period of time or space. This study provides statistical models for two time-series methods: a trend analysis and a seasonal component. Both are within the same time scale. Among the most effective approaches for analysing time series data is the model introduced by Box and Jenkins, called the Autoregressive Integrated Moving Average (ARIMA). The data in Figure 2 were examined to identify the most appropriate class of ARIMA processes. This is achieved by selecting the order of the consecutive and seasonal differencing required to make the series stationary and also specifying the order of the regular and seasonal autoregressive and moving average polynomials necessary to adequately represent the time series model.

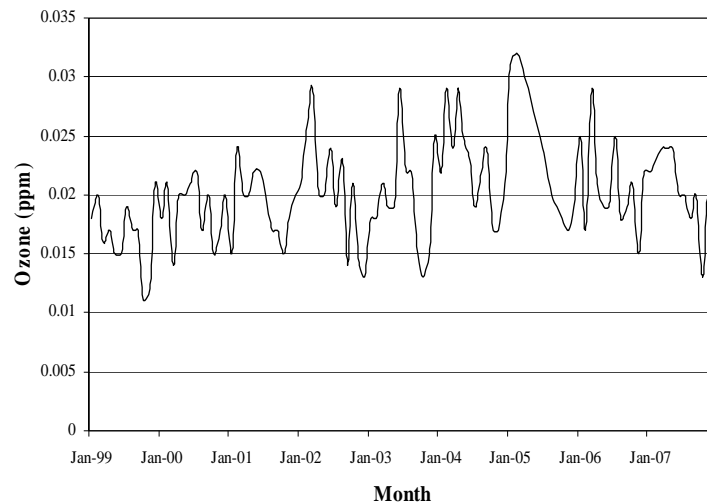


Figure 2: Monthly ozone concentration for Sungai Petani (1999–2007).

2.4 Seasonal Model

Seasonal decomposition was used to decompose the seasonal series into a seasonal component, a combined trend, a cycle component and a short-term variation component, i.e.,

$$O_t = T_t \times S_t \times I_t$$

where O_t is the original ozone time series, T_t the long-term trend component, S_t the seasonal variation, and I_t the short-term variation component, or the error component. As the seasonality increases with the level of the series, a multiplicative model was used to estimate the seasonal index. Under this model, the trend has the same units as the original series, but the seasonal and irregular components are unitless factors that are distributed around 1. As the underlying level of the series changes, the magnitude of the seasonal fluctuations varies as well. The seasonal index was the average deviation of each month's ozone value from the ozone level that was due to the other components in that month.

2.5 Trend Analysis Model

In the trend analysis, the Box-Jenkins ARIMA was applied to model the time series behaviour in generating the forecasting trend. A methodology consisting of a four-step iterative procedure was used in this study.

In the model identification (step 1), the data were examined to check for the most appropriate class of ARIMA processes by: selecting the order of the consecutive and seasonal differencing required to make the series stationary; and specifying the order of the regular and seasonal autoregressive and moving average polynomials necessary to adequately represent the time series model. The Autocorrelation Function (ACF) and the Partial Autocorrelation Function (PACF) are the most important elements of the time series analysis and forecasting. The ACF measures the amount of linear dependence between observations in a time series that is separated by a lag k . The PACF plot helps to determine how many autoregressive terms are necessary to reveal one or more of the following characteristics: time lags where high correlations appear, the seasonality of the series and trends either in the mean level or in the variance of the series. The general model introduced by Box and Jenkins includes autoregressive and moving average parameters as well as differencing in the formulation of the model.

The three types of parameters in the model are the autoregressive parameters (p), the number of differencing passes (d) and moving average

parameters (q). The Box-Jenkins model is summarised as ARIMA (p, d, q). For example, a model described as ARIMA (1,1,1) means that it contains 1 autoregressive (p) parameter and 1 moving average (q) parameter for the time series data after it was differenced once to attain stationarity. In addition to the non-seasonal ARIMA (p, d, q) model introduced above, we could identify seasonal ARIMA (P, D, Q) parameters for our data. These parameters are seasonal autoregressive (P), seasonal differencing (D) and seasonal moving average (Q). Seasonality is defined as a pattern that repeats itself over a fixed interval of time. In general, seasonality can be found by identifying a large autocorrelation coefficient or a large partial autocorrelation coefficient at a seasonal lag. For example, ARIMA (1,1,1)(1,1,1)¹² describes a model that includes 1 autoregressive parameter, 1 moving average parameter, 1 seasonal autoregressive parameter and 1 seasonal moving average parameter. These parameters were computed after the series was differenced once at lag 1 and once at lag 12.

For the seasonal model, we used the Akaike Information Criterion (AIC) for model selection. The AIC is a combination of two conflicting factors: the mean square error and the number of estimated parameters of a model. Generally, the model with the smallest value of AIC is chosen as the best model.¹³

After choosing the most appropriate model, the model parameters are estimated (step 2), and the plot of the ACF and PACF of the stationary data are examined to identify the suggested autoregressive or moving average terms. Here, the values of the parameters are chosen using the least square method to make the Sum of the Squared Residuals (SSR) between the real data and the estimated values as small as possible. In most cases, the nonlinear estimation method is used to estimate the above-identified parameters to maximise the likelihood (probability) of the observed series given the parameter values.¹⁴

In the diagnostic checking step (step 3), the residuals from the fitted model are examined against adequacy. This step is usually done by correlation analysis through the residual ACF plots and the goodness-of-fit test by means of chi-squared statistics divided by 2. If the residuals are correlated, then the model should be refined as in step one above. Otherwise, the autocorrelations are white noise and the model is adequate to represent our time series.

The final stage of the modelling process (step 4) is forecasting, which gives the results as three different options: forecasted values, upper limits and lower limits that provide a confidence interval of 95%. Any forecasted values within the confidence limit are satisfactory. Finally, the accuracy of the model is

checked with the Mean-Square error (MS) to compare fits of different ARIMA models. A lower MS value corresponds to a better fitting model.

3. RESULTS AND DISCUSSION

3.1 Annual Variation of Monthly Means

In the seasonality of the ozone, a well-defined annual cycle was consistent with the highest ozone means occurring in July and the lowest in December (Figure 3). Table 1 shows the seasonal indices for each month scaled so that an average month equals 100. The indices range from a low of 0 in December to a high of 220 in July. The seasonal variation pattern in Sungai Petani differed from other countries such as the United States, the United Kingdom, Italy, Canada, and Japan, in that the peak ozone concentration did not correspond to the maximum photochemical activity in summer.¹⁵

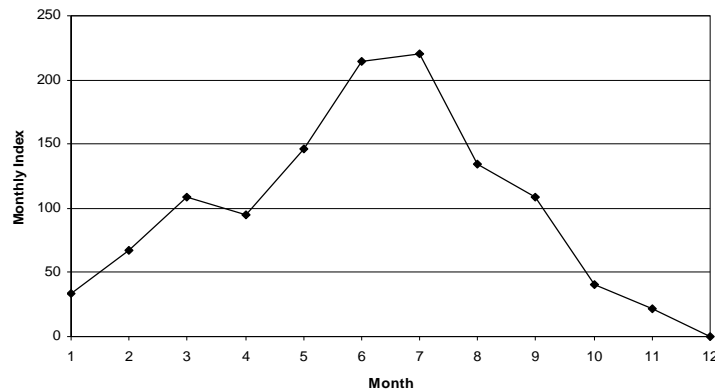


Figure 3: Annual variation of monthly ozone means.

3.2 Trend of Ozone

The model with the lowest value (-11.4628) of the Akaike Information Criterion (AIC) is (ARIMA) $(1, 0, 1) \times (2, 1, 2)$ ¹² with a constant selected and used to generate the forecasts (Figure 4). This model assumes that the best forecast for future data is given by a parametric model relating the most recent data value to previous data values and previous noise. As shown in Table 2, the P-values for the AR (1), MA (1), SAR (1), SAR (2), SMA (1), and SMA (2) terms are less than 0.05, so they are significantly different from 0. Meanwhile, the estimated standard deviation of the input white noise equals 0.00311745.

Since no tests are statistically significant at the 95% or higher confidence level, the current model is adequate to represent the data and can be used to forecast the upcoming ozone concentration.

Table 1: Seasonal index of ozone.

Season	Index
January	33.16
February	67.09
March	108.76
April	94.48
May	145.92
Jun	214.87
July	220.02
August	134.05
September	108.97
October	40.31
November	22.19
December	0

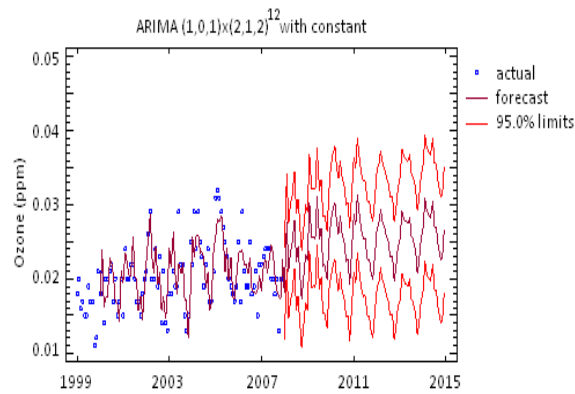


Figure 4: Model predicted plot of ozone concentration with actual and 95% confidence bands.

Table 2: ARIMA model summary.

Parameter	Estimate	Std. Error	T	P-value
AR(1)	0.753937	0.19179	3.93105	0.000167
MA(1)	0.516572	0.246432	2.09621	0.038902
SAR(1)	0.6414	0.0623069	10.2942	0.000000
SAR(2)	-0.62465	0.0504516	-12.3812	0.000000
SMA(1)	1.68401	0.0473854	35.5387	0.000000
SMA(2)	-0.766594	0.041257	-18.581	0.000000
Mean	0.000577368	0.000173008	3.33724	0.001236
Constant	0.000139689			

Estimated white noise variance = 0.00000971852 with 89 degrees of freedom

Estimated white noise standard deviation = 0.00311745

Number of iterations: 16

According to the plots of residual ACF (Figure 5) and PACF (Figure 6), the residuals are white noise and not autocorrelated. Furthermore, as shown in Figure 7 of the normal probability plot, the residuals of the model are normal.

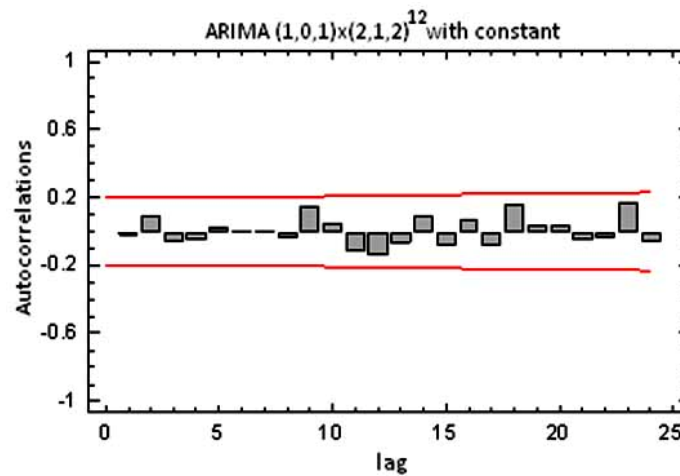


Figure 5: Residual autocorrelation function (ACF) plot.

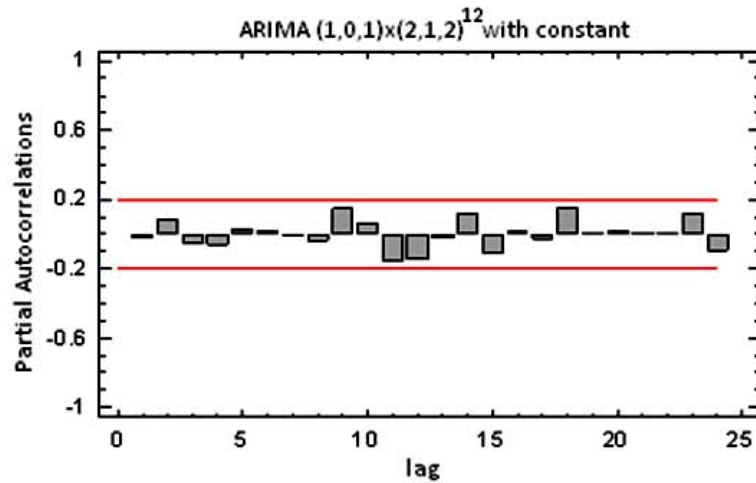


Figure 6: Residual partial autocorrelation function (PACF) plot.

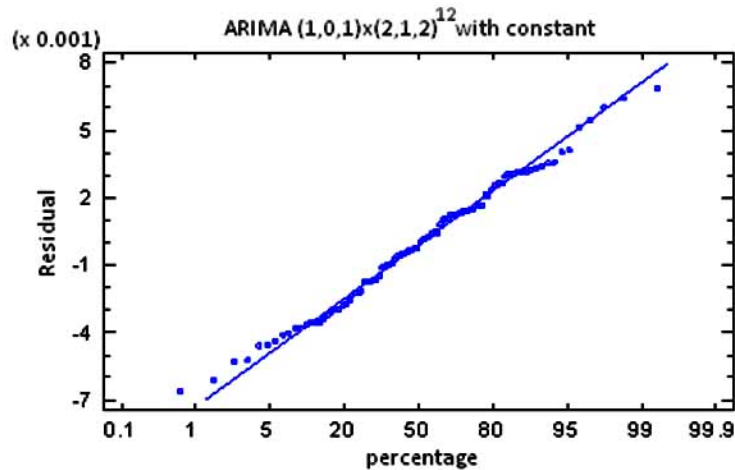


Figure 7: Residual normal probability plot.

Based on the prediction for ozone concentration (Figure 4), there is a statistically significant upward trend at Sungai Petani station. The detection of a steady, statistically significant upward trend of ozone concentration in the MADA area is alarming. This upward trend is likely due to sources of ozone precursors related to industrial activities from nearby areas and the increase in road traffic volume.

3.3 Ozone Exceedances

120 $\mu\text{g}/\text{m}^3$ (60 ppb) was the lowest threshold level selected, as ozone effects can occur at concentrations above this level.¹⁶ This threshold value is in accordance with the mandates of the Malaysian Clean Air Regulation, 1978. We find that the exposure-plant response index [Accumulated exposure Over a Threshold of 40 ppb (AOT40)] and the target values for the protection of human health (8-h > 60 ppb) are regularly surpassed. In fact, exceedance of hourly average concentrations of 60 ppb (120 $\mu\text{g}/\text{m}^3$) are on the rise every year, as shown in Figure 8. This is a cause for concern to agriculture and human health.¹⁷

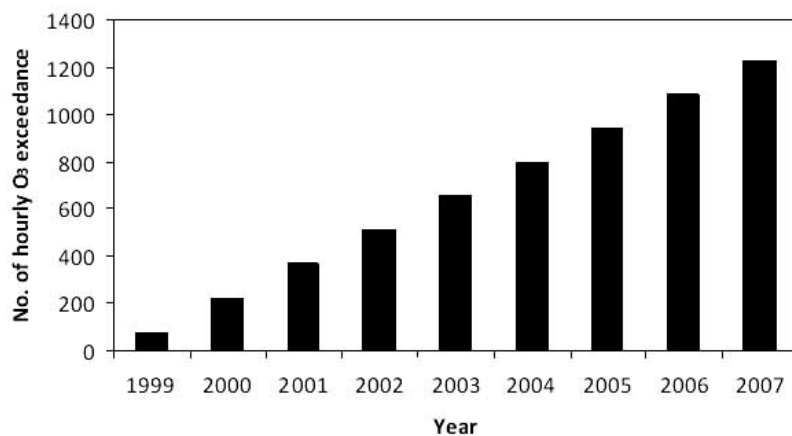


Figure 8: number of hourly O₃ exceedances per year.

4. CONCLUSION

Our analysis shows that, as a whole, concentrations of ground level ozone in the MADA area have been rising steadily, and as ozone is harmful to vegetation, one direct effect of increasing ozone is expected to be a reduction in yield. Furthermore, an increasing number of exceedances of the hourly average concentrations of 60 ppb (120 $\mu\text{g}/\text{m}^3$) is alarming because it will have a negative effect on human health.

5. ACKNOWLEDGEMENTS

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