



Air Quality Time Series Based GARCH Model Analyses of Air Quality Information for a Total Quantity Control District

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ABSTRACT

Air quality data collected at 8 monitoring stations located in the central Taiwan Air Quality Total Quantity Control District were analyzed using multivariate statistical factor analyses. Based on the results thus obtained, 2 major factors, i.e. photochemical pollution factor and fuel factors, were selected for the purpose of evaluating their variations and the pattern of mutual influences for the various air pollution species with respect to time series. The evaluation was conducted using a vector time series coordinated with the ARCH (Autoregressive Conditional Heteroscedacity) and GARCH (Generalized Autoregressive Conditional Heteroscedacity) models in addition to being combined with dynamic impact response analyses using a multiple time series model. The results reveal that the current O₃ value is affected by the PM₁₀ values of both a one time lag and a two times lag, as well as the NO₂ value of one time lag. When the current SO₂ is produced, its concentration can be used to estimate the current CO concentration, and the one time lag SO₂ concentration also influences the CO concentration. Additionally, results of impact response analyses show that current CO concentration responds to variations in current SO₂; this indicates that the existence of SO₂ due to incomplete combustion at the pollution source is immediately reflected by the current production of CO without lagging. In this paper, the vector time series is coupled with the (G)ARCH model to convert simple data series into valuable information so that raw data are better and more completely presented for the purpose of revealing future variation trends. Additionally, the results can be referenced by authorities for planning air quality total quantity control, applying and examining various air quality models, simulating the allowable increase of air quality limits, and evaluating the benefit of air quality improvement.

Keywords: GARCH; Time series; Air quality total quantity control district; Impact response analyses; Air pollution.

INTRODUCTION

The development of (time series) theory, especially for financial series, was pioneered by Box and Jenkins (1976), who proposed the ARIMA (autoregressive integrated moving average) model for performing series (time series), especially financial series analyses. The classic regression analysis model assumes that variables of residual values are constants and that the expected values for every period remain the same. However, the results of many studies have indicated the time-varying nature of sequence order variables. Hence, Engle (1982) proposed the ARCH (autoregressive conditional heteroscedasticity) model that has been further modified by Carson *et al.* (2008) to the System-GARCH Model.

Following Engle's ground breaking idea, many alternatives have been proposed to model conditional variances, forming an immense ARCH family; for example, the survey of Bollerslev *et al.* (1992), and Li *et al.* (2002). Of these models, the most popular is undoubtedly the generalized autoregressive conditional heteroskedasticity (GARCH) model of Bollerslev (1986). Some multivariate extensions of these models have been proposed: for example, Ling and Deng (1993), Engle and Kroner (1995), Wong and Li (1997) and Li *et al.* (2001). In most of these multivariate extensions, the primary purpose has been to investigate the structure of the model, as in Engle and Kroner (1995), and to report empirical findings.

At present, three of the most popular models to capture the time-varying volatility of financial time series are the Generalised Autoregressive Conditional Heteroscedasticity (GARCH) model of Engle (1982) and Bollerslev (1986), the GJR (Glosten, Jagannathan and Runkle) model of Glosten *et al.* (1992), and the Exponential GARCH (EGARCH) model of Nelson (1991). Multivariate extensions of GARCH models are also available in the literature, such as the Constant

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Conditional Correlation (CCC) GARCH model of Bollerslev (1990), the Vector Autoregressive Moving Average GARCH (VARMA-GARCH) model of Ling and McAleer (2003), and the VARMA Asymmetric GARCH (VARMA-AGARCH) model of Hoti *et al.* (2002).

Various studies have also used neural-networks for forecasting air quality (Tsai *et al.*, 2009). Kumar and DeRidder (2010) took heteroskedasticity of the O₃ time series explicitly into account to show how to forecast O₃ with improved confidence intervals. Moreover, their method is capable of making more accurate probability forecasts of ozone episodes in urban areas. Some studies that compare forecasting performances of ARIMA and neural-networks have been carried out by Tang *et al.* (1991), Shabri (2001), Ho *et al.* (2002), and Choon and Chuin (2008) but without a single conclusive result.

Numerous papers have been published on the pollution potential and distribution characteristics of various air pollutants. Zhang *et al.* (2010) has conducted successive measurements and samplings of Asian dust particles in the spring in Beijing since 2001. In this study, the chemical element composition of aerosol particles was characterized through the ground-based samples of size-segregated aerosols collected in Beijing during dust events in the spring of 2004. Huboyo *et al.* (2011) investigated the characteristics of fine particles (PM_{2.5}) associated with cooking, particularly temporal variations in the mass and number concentrations in a kitchen and an adjoining room. Chou *et al.* (2006) have studied the relationships among ambient levels of O₃, NO and NO₂ to improve the understanding of the chemical coupling occurring among them. Cheng and Lin (2010) showed that the low PM_{2.5}-to-PM₁₀ ratio at the concourse in the Taipei main station was likely the result of coarse PM being re-suspended in the station concourse due to passenger movement. Additionally, Han *et al.* (2011) used O₃, NO and NO₂ concentrations to forecast the O₃ concentration. Furthermore, the difference in concentrations between the weekdays and weekends, and O₃ concentrations under different meteorological conditions were also discussed.

In recent years, the increasing number of industrial plants, automobiles, and motorcycles in Taiwan has made it difficult to improve the air quality of regions with concentrated sources of pollution regardless of regulations implementing more stringent air quality. Hence, promoting an air quality total quantity control is a strategy for further improving the air quality in Taiwan. The Environmental Protection Administration (Taiwan) divides Taiwan into seven air quality administration districts: Northern Taiwan District, Hsinchu-Miaoli District, Central Taiwan District, Yunlin-Chiayi-Tainan District, Kaohsiung-Pingtung District, Hualien-Taitung District, and Yilan District. Based on actual situations and requirements, the air quality total quantity limitations will be phased in for each district. The first plan to reduce the air quality total quantity will be implemented in two districts that have the worst air quality total quantity, i.e. the Central Taiwan District and the Kaohsiung-Pingtung Districts. Therefore, the Central Taiwan District, which includes metropolitan Taichung, Changhua County and Nantou County, were selected for the purpose

of conducting this study using the data collected by 8 air quality monitoring stations established by the Environmental Protection Administration (Taiwan) in these districts in order to carry out a multivariate statistical factor analyses. Based on the results, the air quality situations were categorized into three factors, and the two most important factors, i.e. the photochemical pollution factor and the fuel factor, were targeted for further analyses. Because dynamic variations exist among the various air pollution species, the second moment information needs to be completely grasped so that the tendency of time-dependent variations can be analyzed using the ARMA-GARCH model. Hence, patterns and results of the mutual influence among the various air pollution species can be investigated. The results will instantaneously reflect the response and correlation of various air pollution parameters and can be referenced by the government for review and certification of various air quality models in order for agencies to simulate allowable increase limits and evaluate the benefits of air quality improvement.

METHODS

Selection of Air Quality Monitoring Stations

The Taichung Thermal Power Plant located in this study region was completed in 1989; it is the largest CO₂ emitter in Taiwan. With the development of the Changhua Coastal Industrial Park in this region, the air quality of the whole central region has been made worse. According to monitored air quality data collected by the Environmental Protection Administration (Taiwan), the air quality in this region is categorized in the third air quality protection class with PM₁₀ (particulate matter with particle size below 10 microns) and O₃ (ozone) seriously exceeding the air quality standards. The 8 ordinary air quality monitoring stations established by the Environmental Protection Administration (Taiwan) include: Xitun Station, Fengyuan Station, Shalu Station, Dali Station in municipal Taichung, Changhua Station, Erlin Station in Changhua County, Nantou Station, and Zhushan Station in Nantou County. Fig. 1 shows the geographical locations of these stations in the central area of Taiwan.

Screening and Manipulation of Data

Prior to performing GARCH simulation studies, the air quality data collected by the 8 air quality monitoring stations were first analyzed using multivariate statistical factor analyses to look for common factors and to investigate the mutual correlations among the 7 air pollution species. In addition to the 5 major air pollutants Standard Index (PSI) as published by Environmental Protection Administration (Taiwan), including sulfur dioxide (SO₂), nitrogen dioxide (NO₂), carbon monoxide (CO), PM₁₀ and ozone (O₃), two more indices, i.e. total hydrocarbon compounds (THC) and non-methane hydrocarbon compounds (NMHC) were added in this study. The air quality index (AQI) proposed by US EPA uses the pollution potential of these 5 major air pollutants to classify the degree of their influence on human health. In literature, many authors have investigated the distribution of these 5 major air pollutants in the atmosphere (Bhaskar and Mehta, 2010; Hussein *et al.*, 2011). Using all



Fig. 1. Geographic locations of the 8 air quality monitoring stations established by the Environmental Protection Administration (Taiwan) in the study region.

7 of these indices will ensure the completeness and variety of the air quality data used in the subsequent analyses. Results of factor analyses show that three factors have eigenvalues greater than “1”; they are the organic pollution factor (i.e. NMHC, THC), the photochemical pollution factor (NO_2 , PM_{10} , O_3) and the fuel factor (i.e. SO_2 , CO). Among the above 3 factors, NO_2 , PM_{10} and O_3 , which represent the photochemical pollution factor, are subjected to multivariate statistical factor analyses. The loading degrees are 0.866 for NO_2 , 0.751 for PM_{10} , and 0.691 for O_3 , whereas the loading degrees for all other air-borne pollutants are not high. In the case of SO_2 and CO , which are represented in the fuel factor, the factor loading degrees are 0.872 and 0.754, respectively, and all other factors in the fuel factor are not high either. If some air pollutants in one factor have relatively high factor loading degrees, these pollutants have relatively high dependence among one another. Only the photochemical pollution factor and fuel factor were used to carry out the GARCH model simulation because the air pollutants included in these two factors cover the 5 air pollutants in the PSI published by Environmental Protection Administration (Taiwan). It can be surmised that using these two factors will fully represent the air quality pollution

situation in the Central Taiwan District. The two air pollutants included in the organic pollution factor do not contribute significantly to air pollution problems in this district; they are not stipulated by the Taiwan Environmental Protection Administration as currently evaluated air pollutant standard indices either. Hence, further discussions on these two air pollutants are not included in this paper.

The series of data used in this study consist of 610 sets collected daily by the Environmental Protection Administration (Taiwan) from January 1, 2010 to September 30, 2011, and published in <http://www.epa.gov.tw>. During this period, some data were incompletely collected because of un-expected instrument down time for repair and maintenance, and all of these incomplete data sets are deleted, resulting in 610 complete sets of data. All statistical analyses were carried out with E-Views for Windows, version 6.0.

ARIMA Modeling (Shumway and Stoffer, 2006)

A time series $\{x_t; t = 0, \pm 1, \pm 2, \dots\}$ is ARMA(p, q) if it is covariance stationary and can be represented as:

$$x_t = \phi_1 x_{t-1} + \dots + \phi_p x_{t-p} + \varepsilon_t + \theta_1 \varepsilon_{t-1} + \dots + \theta_q \varepsilon_{t-q}, \quad (1)$$

where $\phi_p \neq 0$, $\theta_q \neq 0$, and ε_t are the innovations with $N(0, \sigma_\varepsilon^2)$, and $\sigma_\varepsilon^2 > 0$.) The parameters p and q are called the autoregressive [AR(p)] and the moving average [MA(q)] orders, respectively. When a time series does not appear to be covariance stationary, the differencing procedure may be applied to make it stationary. Subsequently, the ARMA(p, q) model can be applied to the stationary differenced time series; the model so constructed is called the ARIMA(p, d, q) model, where d denotes the order of differencing (Shumway and Stoffer, 2006; Brockwell and Davis, 2002). Parameters ϕ and θ are estimated using the maximum likelihood method (Brockwell and Davis, 2002) in this study.

An inspection of both the autocorrelation function (ACF) and the partial autocorrelation function (PACF) assists in identifying the orders AR(p) and MA(q). In addition, more objectively defined criteria, such as the Akaike information criterion (AIC), the Hannone-Quinn Information Criterion (HIC), the Bayesian Information Criterion (BIC) and the Final Prediction Error (FPE) can also be used to identify the correct of p and q (Brockwell and Davis, 2002; Kumar *et al.*, 2009).

GARCH Modeling (Kumar and De Ridder, 2010)

If values for ε_t denote a real valued discrete-time stochastic process, ε_t are the innovations of the ARMA process in Eq. (1). Engle (1982) defined these parameters as an autoregressive conditional heteroskedastic process in which all ε_t are expressed as:

$$\varepsilon_t = z_t \sigma_t, \tag{2}$$

where z_t is an identically independent distributed process with a zero mean and unit variance. Although values for ε_t are serially uncorrelated by definition, their conditional variances equal σ_t^2 , which might be autocorrelated and, therefore, may change over time.

The variance equation of the GARCH (p, q) can be expressed as (Bollerslev, 1986; Aradhyula and Holt, 1988; Shumway and Stoffer, 2006; Brockwell and Davis, 2002):

$$z_t \sim D_\theta(0, 1) \tag{3}$$

$$\sigma_t^2 = \alpha_0 + \sum_{i=1}^q \alpha_i \varepsilon_{t-i}^2 + \sum_{i=1}^p \beta_i \sigma_{t-i}^2 \tag{4}$$

$$\sigma_t^2 = \alpha_0 + \alpha(B)\varepsilon_{t-1}^2 + \beta(B)\sigma_{t-1}^2, \tag{5}$$

where $\alpha(B)$ and $\beta(B)$ are the appropriate polynomials of the lag operator B ; $D_\theta(0, 1)$ is the probability density function of the innovations or residuals with zero mean and unit variance;

$$p \geq 0; q \geq 0 \tag{6}$$

$$\alpha_0 > 0, \alpha_i \geq 0, i = 1, 2, \dots, q, \text{ and} \tag{7}$$

$$\beta_i \geq 0, i = 1, 2, \dots, p. \tag{8}$$

If p equals 0, the process reduces to an ARCH(q) process.

Also, if both p and q equal 0, the conditional variance is constant. As in ARMA, the innovation ε_t simply reduces to white noise.

VARMA-GARCH Modeling

Serletis and Shahmoradi (2006) proposed an extended version of the VARMA (vector autoregressive moving average)-GARCH model to simulate natural gas price changes (g_t), and electricity price changes (e_t). The model is expressed as follows:

$$y_t = a + b\hat{\varepsilon}_{t-1} + \sum_{i=1}^p \Gamma_i y_{t-i} + \sum_{j=0}^q \Psi_j h_{t-j} + \sum_{k=1}^r \Phi_k z_{t-k} + \sum_{l=1}^s \Theta_l u_{t-l} + u_t \tag{9}$$

with

$$u_t | \Omega_{t-1} \sim (0, H_t), \quad H_t = \begin{bmatrix} h_{gt} & h_{ge_t} \\ h_{ge_t} & h_{e_t} \end{bmatrix},$$

where Ω_{t-1} denotes the available information set in period $t-1$, and

$$y_t = \begin{bmatrix} g_t \\ e_t \end{bmatrix}; \quad u_t = \begin{bmatrix} u_{gt} \\ u_{e_t} \end{bmatrix}; \quad h_t = \begin{bmatrix} h_{gt} \\ h_{e_t} \end{bmatrix}; \quad a = \begin{bmatrix} a_g \\ a_e \end{bmatrix};$$

$$\Gamma_t = \begin{bmatrix} \gamma_{11}^{(i)} & \gamma_{12}^{(i)} \\ \gamma_{21}^{(i)} & \gamma_{22}^{(i)} \end{bmatrix}; \quad \Psi_t = \begin{bmatrix} \Psi_{11}^{(j)} & \Psi_{12}^{(j)} \\ \Psi_{21}^{(j)} & \Psi_{22}^{(j)} \end{bmatrix};$$

$$\Phi_k = \begin{bmatrix} \Phi_{11}^{(k)} & \Phi_{12}^{(k)} \\ \Phi_{21}^{(k)} & \Phi_{22}^{(k)} \end{bmatrix}; \quad \Theta_l = \begin{bmatrix} \theta_{11}^{(l)} & \theta_{12}^{(l)} \\ \theta_{21}^{(l)} & \theta_{22}^{(l)} \end{bmatrix};$$

$$z_{t-k} = \begin{bmatrix} z_{gt-k} \\ z_{e_t-k} \end{bmatrix}; \quad z_{jt-k} = \frac{u_{jt-k}}{\sqrt{h_{jt-k}}}, \text{ for } j = g, e.$$

Parameters h_{t-j} and z_{t-k} are introduced to take the anticipated and unanticipated volatilities into account. $\hat{\varepsilon}_{t-1}$ is the error correction term from the long run cointegrating regression.

Impact response Analyses Functions

An impulse response function measures the time profile of the effect of shocks at a given point in time on the (expected) future values of variables in a dynamic system. The augmented vector autoregressive model is used for carrying out the impact response analyses:

$$X_t = \sum_{i=1}^p \Phi_i X_{t-i} + \Psi w_t + \varepsilon_t, \quad t = 1, 2, \dots, T, \tag{10}$$

where $X_t = (x_{1t}, x_{2t}, \dots, x_{mt})'$ is an $m \times 1$ vector of jointly determined dependent variables; w_t is a $q \times 1$ vector of deterministic and/or exogenous variables; and $\{\Phi_i = 1, 2, \dots, p\}$, and Ψ are $m \times m$ and $m \times q$ coefficient matrices. The following standard assumptions are made (Lütkepohl, 1991):

Assumption 1: $E(\varepsilon_t) = 0, E(\varepsilon_t \varepsilon_t') = \Sigma$ for all t , where $\Sigma =$

$\{\sigma_{ij}, i, j = 1, 2, \dots, m\}$ is an $m \times m$ positive definite matrix, $E(\varepsilon_t \varepsilon_t') = 0$ for all $t = t'$ and $E(\varepsilon_t | w_t) = 0$.

Assumption 2: All the roots of $|I_m - \sum_{i=1}^p \Phi_i z^i| = 0$ fall outside the unit circle.

Assumption 3: $X_{t-1}, X_{t-2}, \dots, X_{t-p}, w_t, t = 1, 2, \dots, T$, are not perfectly collinear.

Under Assumption 3, X_t would be covariance-stationary, and Eq. (10) can be rewritten as the infinite moving average representation:

$$X_t = \sum_{i=0}^{\infty} A_i \varepsilon_{t-i} + \sum_{i=0}^{\infty} G_i w_{t-i}, \quad t = 1, 2, \dots, T, \quad (11)$$

where the $m \times m$ coefficient matrices A_i can be obtained using the following recursive relations:

$$A_i = \Phi_1 A_{i-1} + \Phi_2 A_{i-2} + \dots + \Phi_p A_{i-p}, \quad i = 1, 2, \dots \quad (12)$$

with $A_0 = I_m$ and $A_i = 0$ for $i < 0$, and $G_i = A_i \Psi$.

Essence of Model Development

The model used to simulate and predict the two selected factors was calibrated using statistical principles and methods in order to expound the significance of the fat tail test, the Ljung-Box series examination, and the ARCH examination.

Fat Tail Test

The investigation of time series empirical distribution often leads to characteristics of the fat tail test. Hence, the assumption of a normal distribution for the air quality data is not the optimal choice. An examination of the results of the skewness, kurtosis, and the Jarque-Bera normal distribution test can be used to determine whether the distribution of modeling errors has fat tails.

Ljung-Box Series Examination

The residual needs to be examined to determine whether it shows a sequential correlation before evaluating the ARCH and GARCH models. The square of a residual that shows sequential correlations will have the ARCH effect.

Examination of the ARCH Effectiveness

Before conducting simulations using the time series combination of the ARCH and GARCH models, the process of model calibration must be carried out beforehand in order to confirm that the residual series is not related to the first order series, known as white noise, to assure an appropriate model. Next, the residual square examination is used to determine whether the model has the (G)ARCH effect. In this paper, the Q statistics proposed by Ljung-Box are used to examine whether the residual has high order autocorrelation. Only a model that has ARCH effectiveness can be used to carry out iterative non-linear calculations for estimating model parameters.

RESULTS AND DISCUSSION

Evaluating the Photochemical Pollution Factor Simulation Model

Photochemical pollution factors include NO₂, O₃, and PM₁₀.

Analyses of Basic Characteristics

Table 1 lists the basic characteristics of these 3 air pollutants, including average, standard deviation, skewness, kurtosis, and the Jarque-Bera normal distribution examination statistics. All data show the phenomenon of “skewed on right” because they have positive skewness; NO₂ that has the highest skewness (4.86) indicates that several NO₂ data in this series experience the phenomenon of sudden increase. In contrast, PM₁₀ has a relatively low skewness of only 0.86 because the central air quality total quantity control district is located near two major air pollution sources, i.e. the Taichung Thermal Power Plants and the Changhua Coastal Industrial Park, causing higher PM₁₀ concentration and the resulting air pollution problem. This district has a relative higher PM₁₀ concentration during winter, so the skewness is not quite significant. The O₃ skewness of 3.89 also indicates that O₃ is a major species that causes air pollution; however, the number of days for relatively higher O₃ is relatively lower than that for relatively higher PM₁₀ as indicated by the relatively higher PM₁₀ skewness as opposed to that of O₃. As for NO₂, the original data indicate that it is higher only during specific periods in winter and early spring; its contribution to air quality problem is relatively insignificant, as shown by its high skewness. All three pollution factors have kurtosis values greater than the kurtosis value of 3 for a normal distribution; hence, the data series have the characteristics of seasonal series. Additionally, the statistical results from examining the Jarque-Bera normal distribution show that the 5% significance level is greater than the critical value (degree of freedom = 2 and $\chi_{0.05,2}^2 = 5.99$). This observation rejects the hypothesis of normal distribution. All three air pollution factors show the characteristics of double fat-tails, indicating that the data series is actually influenced by seasons.

Ljung-Box Series Examination

Results of using the Ljung-Box method to perform series examinations are listed in Table 2. All examination statistical values for L-B-Q(K) are smaller than the critical value so that the critical value so that null hypothesis, which does not conform to the alternative hypothesis, and therefore, cannot be rejected. This indicates that series residuals conform to white noise because they do not show serial correlations, indicating that the model disposition is quite adequate.

Examination of the ARCH Effect

Whether the ARCH effect exists in a series can be examined using the LM (Lagrange Multiplier) statistical quantity, which is expressed as LM, in which T is the number of samples, and R² is the determination coefficient of the results obtained by using the OLS (Ordinary Least Squares) regression. T × R² obeys the chi-square distribution that has a P degree of freedom. When the LM statistical value is

Table 1. Basic statistical characteristics of the various photochemical pollution factors.

	NO ₂ (ppb)	O ₃ (ppb)	PM ₁₀ (µg/m ³)
Mean	58.68113	32.78390	119.6294
Median	56.16000	28.61000	115.7270
Maximum	497.9000	398.2500	458.1804
Minimum	0.150000	0.150000	1.023000
Std. Dev.	47.70383	27.33519	55.85025
Skewness	4.860983	3.896530	0.864264
Kurtosis	38.50992	37.17446	4.89193
Jarque-Bera	140196.8	136704.4	634.2893
Probability	0.000000	0.000000	0.000000
Sum Sq. Dev.	4778877.	1569147.	6550427.
Observations	610	610	610

Table 2. Determination of Ljung-Box series correlation for the various photochemical pollution factors.

L-BQ(K)	NO ₂	O ₃	PM ₁₀	Critical value	$\chi^2_{(0.05,k)}$
1	0.68	0.32	0.28	3.84	
2	1.74	1.06	1.33	5.99	
3	4.01	2.50	2.86	7.82	
4	5.43	4.90	4.01	9.49	
5	6.67	6.72	5.42	11.07	
6	7.19	7.90	5.98	12.59	
7	7.72	9.12	7.17	14.07	
8	8.11	10.03	7.86	15.51	
9	9.54	10.74	8.55	16.92	
10	10.92	12.77	10.08	18.31	
16	14.55	16.58	12.91	26.30	
20	22.18	23.14	16.80	31.41	
24	25.17	27.53	23.22	36.42	

Note: $R_t = c + \theta R_{t-1} + \varepsilon_t$; $\alpha = 0.05$.

higher than the 5% significance level, the series shows the ARCH effect. The statistical results of the three selected air pollutants shown in Table 3 reveal that all $T \times R^2$ values are greater than the 5% significance level and that the conditional variances of all three parameters have a strong ARCH effect. Hence, using the ARCH effect to explain the photochemical pollution factor is quite appropriate.

Selecting ARCH and GARCH Models

The vector model EACF paired with various combinations of the ARCH and GARCH models has been tested in order to select the most appropriate VARMA(p,d,q)-GARCH(p,q) model for carrying out the simulation analyses. Table 4 lists the results; the VARMA(2,0,1)-GARCH(2,1) combination is selected because it has the lowest AIC and SC values. Table 5 shows the evaluation of parameters used in the VARCH(2,0,1)-GARCH(2,1) model.

Simulation Results

Table 5 shows the resulting equations and other relevant information obtained by carrying out the VARCH(2,0,1)-GARCH(2,1) model simulation. It indicates that when the current PM₁₀ is produced, the current O₃ concentration cannot be estimated based on the PM₁₀ concentration because the b₀ t-statistic of 1.29 is less than 1.96, indicating a lack of significance. However, the one time lag and two times

Lag PM₁₀ concentrations do appear to influence the formation of O₃ concentrations. The t-statistic values of b₁ for the one time lag PM₁₀ (2.56), and b₂ for the two times lag PM₁₀ (1.99) are greater than 1.96; both indicating significance. Although some environmental engineering textbooks suggest that the production of PM₁₀ is not directly related to the production of O₃, Chou (2010) has pointed out that the secondary aerosols of photochemical reactions cause high levels of atmospheric PM_{2.5} and PM₁₀ in Taiwan. Additionally, atmospheric PM₁₀ includes primary and secondary aerosols, and the secondary aerosols may be classified as either secondary inorganic aerosols or secondary organic aerosols. Chen and Lee (1999) and Chang and Lee (2007) have observed that secondary organic aerosols and photochemical reactions of VOCs are closely related to the formation of atmospheric O₃. Based on these statements, the production of PM₁₀ should be closely related to the photochemical reactions characteristic of atmospheric pollutants. In this research, the model simulation results also reveal that the one lag time atmospheric PM₁₀ concentration is actually related to the prediction of O₃ concentration. Thus, the relationship between atmospheric PM₁₀ concentration and O₃ production has been confirmed by measured and simulated data. As for NO₂, its current concentration cannot be used to predict the current O₃ concentration (c₀ t-statistic of 1.00 is less than 1.96, indicating a lack of significance). However,

Table 3. Results of ARCH(q) effect examination for the various photochemical pollution factors.

Q (lagged variables)	NO ₂ (TR ²)	O ₃ (TR ²)	PM ₁₀ (TR ²)	Critical value	$x^2_{(0.05,k)}$
1	147.04	6.31	547.88	3.84	
2	193.42	8.22	570.86	5.99	
3	196.35	10.70	575.10	7.82	
4	198.40	21.17	625.11	9.49	
5	208.36	23.92	630.08	11.07	
6	247.73	23.01	635.77	12.59	
7	290.55	24.98	633.39	14.07	
8	329.81	24.74	636.50	15.51	
9	340.02	28.86	639.23	16.92	
10	380.57	30.03	637.62	19.68	

Note: All $T \times R^2$ values are greater than 5%, indicating "significance".

Table 4. Results of VARMA-GARCH examination for the various photochemical pollution factors.

VARMA	GARCH	ARCH(1)		ARCH(2)		GARCH(1,1)		GARCH(2,1)	
		AIC	SC	AIC	SC	AIC	SC	AIC	SC
VARMA(1,0,0)		9.084	9.099	9.048	9.079	9.059	9.078	9.062	9.073
VARMA(2,0,0)		9.056	9.075	9.046	9.070	9.044	9.057	9.012	9.035
VARMA(0,0,1)		9.148	9.159	9.143	9.162	9.145	9.164	9.146	9.167
VARMA(1,0,1)		8.988	9.007	8.978	9.003	8.971	8.990	8.972	8.986
VARMA(2,0,1)		8,991	9.017	8.975	8.989	8.963	8.992	8.943	8.970

Table 5. Parameters obtained using the combined vector model and GARCH(2,1) models for the various photochemical pollution factors.

	a ₀	a ₁	a ₂	b ₀	b ₁	b ₂	c ₀	c ₁	c ₂	d ₁	α ₀	α ₁	α ₂	β ₁
VARMA(1,0,0)	1.61	0.65		0.05	0.17		0.01	-0.12			11.98	0.17	0.21	0.38
t-statistic	14.73	0.38		4.94	1.23		1.20	0.79			16.66	5.96	1.23	-0.16
VARMA(2,0,0)	1.91	0.43	-0.32	0.05	1.43	0.99	0.01	1.52	-1.33		6.00	0.12	0.52	0.31
t-statistic	0.38	-1.45	0.56	6.98	-0.08	1.09	1.29	-1.00	2.31		28.7	0.32	0.75	12.4
VARMA(0,0,1)	1.05			0.10			0.02			0.34	4.4	0.38	-0.35	0.93
t-statistic	0.33			0.54			0.02			22.9	7.16	27.39	-22.0	1.39
VARMA(1,0,1)	2.65	0.96		0.05	0.43		0.01	2.51		-0.7	6.68	0.25	0.49	0.26
t-statistic	9.45	-1.56		6.97	1.02		1.20	-1.87		-4.42	25.43	5.40	1.70	-1.12
VARMA(2,0,1)	3.30	1.18	-0.19	0.05	1.02	0.53	-0.01	0.36	0.51		6.01	0.23	0.52	0.27
t-statistic	7.21	37.5	-0.87	1.29	2.56	1.99	1.00	2.05	1.45		27.86	4.57	11.41	11.50

$$O_3 = a_0 + a_1O_{3(t-1)} + a_2O_{3(t-2)} + b_0PM_{10(t)} + b_1PM_{10(t-1)} + b_2PM_{10(t-2)} + c_0NO_{2(t)} + c_1NO_{2(t-1)} + c_2NO_{2(t-2)} + d_1\varepsilon_{t-1}$$

$$h_t = \alpha_0 + \alpha_1 \varepsilon_{t-1}^2 + \alpha_2 \varepsilon_{t-2}^2 + \beta_1 h_{t-1}$$

the one time lag NO₂ concentration influences the formation of O₃ (c₁ t-statistic of 2.05 is greater than 1.96, indicating significance), whereas the two times lag NO₂ concentration becomes insignificant (c₂ t-statistic of 1.45 is less than 1.96, indicating a lack of significance). The above analyses indicate that the current O₃ concentration is affected by the one time lag and two times lag PM₁₀ concentrations as well as the one time lag NO₂ concentration. A possible explanation for this is that the emission of PM₁₀ and NO₂ to the atmosphere after they are generated by polluting activities will not lead to immediate photochemical reactions; they are subject to a photochemical reaction in order to produce O₃ only during the one time lag and two times lag. PM₁₀ may stay in the atmosphere for a long time (Chou, 2010); however, through photochemical reaction mechanisms, it will cause the production of O₃ even after the two times lag. Hence, the

proceeding of photochemical reactions behind the time when the pollutants are emitted into atmosphere by one time lag or even two times lag. The two times lag PM₁₀ concentration is capable of affecting the formation of the current O₃, and the one time lag NO₂ concentration will affect the formation of current O₃. O₃ concentration is influenced by its own one time lag concentration (the a₁ t-statistic of 37.5 is greater than 1.96, indicating significance), but it is not quite influenced by its own two times lag concentration (the a₂ t-statistic of -0.87 is less than 1.96, indicating a lack of significance). Hence, the current O₃ concentration is influenced by its own one time lag concentration but not its own two times lag concentration.

Analyses of Impact Responses

The AIC (Akaike Information Criterion) proposed by

Akaike (Posada and Buckley, 2004) is used in this paper for selecting appropriate lagged variables:

$$AIC(m) = T \times \ln(SSR/T) + 2m,$$

where m is the number of variables in a model; T is the number of samples; SSR is the sum of error squares.

The AIC of lagged variables are first tested in order to select the one with the smallest AIC value as the most appropriate lagged variable to be used as the basis for subsequent analyses. The results of analyzing lagged variables listed in Table 6 show that the lagged variables of the eighth period have the smallest AIC, so these lagged variables are selected for carrying the impact response analyses in this study. Fig. 2 shows the responses to one-unit variations of the two photochemical air pollution parameters. Mark “1” on the X-axis represents “current”, and mark “2” represent “lag one time”. The Y-axis represents the degree of influence by the production or concentration change of an air pollutant on the “current” or “lag one time” concentration of another air pollutant.

Fig. 2(A) shows responses of PM_{10} and NO_2 to variations in O_3 . When O_3 varies, the current PM_{10} and NO_2 show responses; this indicates that the involvement of PM_{10} and NO_2 in photochemical reactions will lead to the production of O_3 , so the O_3 concentration is influenced by PM_{10} and NO_2 . Therefore, the variation of O_3 concentration can be

Table 6. AIC values of the lagged variables for the various photochemical pollution factors.

Lag items	AIC
1	-3.56
2	-4.03
3	-4.21
4	-3.98
5	-4.43
6	-3.90
7	-4.47
8	-4.73*
9	-3.92
10	-4.08

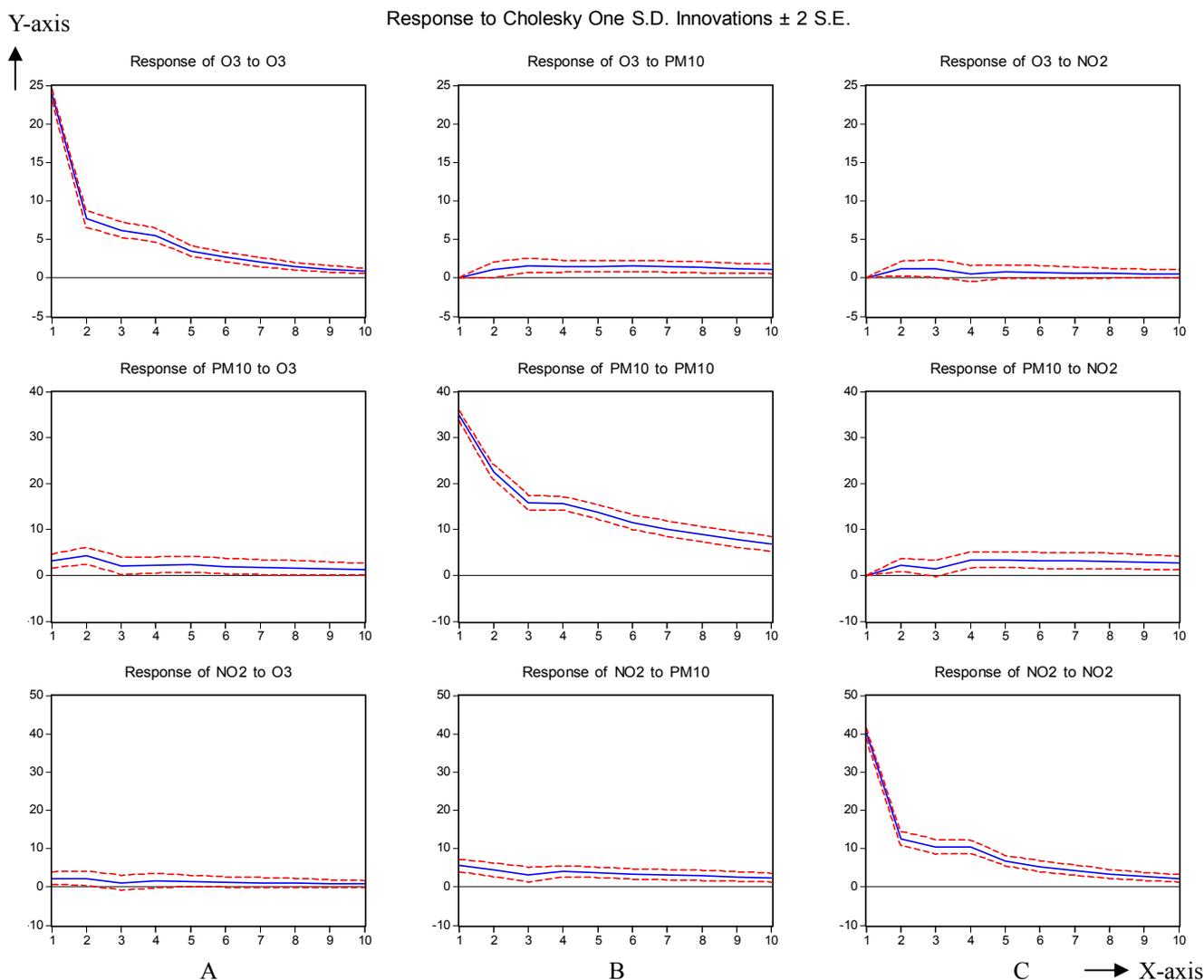


Fig. 2. Analyses of impact response for the various photochemical pollution factors.

generally determined based on concentrations of PM₁₀ and NO₂. As for O₃, it is affected by its own one time lag concentration; hence the current O₃ shows response when the current variation occurs; the influence of the two times lag becomes less and less over time.

Responses of O₃ and NO₂ to variations in PM₁₀ are shown in Fig. 2(B). The current O₃ does not show any response to the initial variation of current PM₁₀ until after the one time lag; this is similar to the simulated results. As discussed in the above simulation results section, secondary aerosols produced by photochemical reactions cause high PM_{2.5} and PM₁₀ levels in the atmosphere of Taiwan. Additionally, the one lag time PM₁₀ is sufficient to predict and influence the atmospheric ozone concentration. Afterward, the O₃ concentration is affected by PM₁₀. Fig. 2(B) also show significant responses of current PM₁₀ to its one lag time; the current PM₁₀ responds immediately to the production of the current PM₁₀.

Fig. 2(C) showing the responses of O₃ and PM₁₀ to variations in NO₂ indicates that the current O₃ or PM₁₀ does not have responses to the variations of current NO₂. When NO₂ is produced by polluting activities, the current O₃ concentration cannot be estimated based on NO₂ concentration until after a one time lag. The figure also reveals that NO₂ has significant response to its own lag time; the current NO₂ responds immediately to variations in its own lag period.

Investigating the Simulation of Fuel Factor

The fuel factors include two air pollutants, i.e. SO₂ and CO.

Analyses of Basic Characteristics

The basic characteristics of statistical parameters such as average, standard deviation, skewness and the Jarque-Bera normal distribution of these two air pollutants are listed in Table 7. All skewness values are positive so that they skew on the right, indicating that several data in the series show the phenomenon of sudden increase. The presence of high PM₁₀ and O₃ in the atmosphere is the major cause of deteriorating air quality, whereas high concentrations of SO₂ and CO cause very few days of deteriorating air quality problems. Hence, unlike the skewness of 0.86 for PM₁₀, the listed skewness of 3.89 for O₃ is somewhat low. In this research, the skewness values are found to be 5.71 for SO₂, and 17.94 for CO. The kurtosis values are 36.82 for SO₂ and 568.98 for CO; both values are greater the maximum coefficient of 3 for a normal distribution. This indicates that these two air pollutants have characteristics of time-dependent series. As for the statistics from the Jarque-Bera normal distribution examination, the 5% significance level for both air pollutants are greater than the critical value, with a degree of freedom of 2 ($\chi^2_{0.05,2} = 5.99$). Hence, the hypothesis that these two series reject the normal distribution indicates the characteristics of double fat-tail. In other words, concentrations of both SO₂ and CO are significantly influenced by seasons in addition to being time-dependent so that different concentrations are observed.

Table 7. Basic statistical characteristics of the various fuel factors.

	SO ₂ (ppb)	CO (ppm)
Mean	26.32675	1.32490
Median	20.87646	1.27805
Maximum	330.8790	27.76435
Minimum	0.176587	0.01
Std. Dev.	23.76512	0.77051
Skewness	5.711058	17.94632
Kurtosis	36.82414	568.9841
Jarque-Bera	111964.6	273049.13
Probability	0.000000	0.000000
Sum Sq. Dev.	1268060	1183.646
Observations	610	610

Examination of the Ljung-Box Series

The results of the Ljung-Box Examination shown in Table 8 reveal that all L-B-Q(K) examination results are smaller than the critical values. Hence, the null hypothesis cannot be rejected, indicating that the residual for either series does not have sequential correlations. This observation conforms to the phenomenon of white noise so that the model disposition is quite appropriate.

Examination of ARCH Effect

Table 9 shows the results of examining the ARCH effect; both the conditional variance values for SO₂ and CO show the ARCH effect as seen by all $T \times R^2$ values being less than the 5% significance level that indicates significance. Hence, the ARCH effect is appropriate for explaining the fuel factor.

Selecting the ARCH and GARCH Models

Table 10 lists the testing results of using the EACF vector model combined with various combinations of the ARCH and GARCH models for evaluating the fuel factors in order to select the most appropriate combination of models. Both the

Table 8. Determination of Ljung-Box series correlation for the various fuel factors.

L-BQ(K)	SO ₂	CO	Critical value $\chi^2_{(0.05,k)}$
1	0.26	0.78	3.84
2	0.74	0.90	5.99
3	2.33	1.23	7.82
4	5.00	3.34	9.49
5	5.97	5.09	11.07
6	7.12	6.37	12.59
7	7.80	8.06	14.07
8	8.87	10.02	15.51
9	10.14	11.28	16.92
10	13.92	12.98	18.31
16	15.16	16.11	26.30
20	21.58	19.95	31.41
24	25.21	22.99	36.42

Note: $R_t = c + \theta R_{t-1} + \epsilon_t$; $\alpha = 0.05$.

Table 9. Results of ARCH(q) effect examination for the various fuel factors.

Q (lagged variables)	SO ₂ (T × R ²)	CO (TR ²)	Critical value x ² _(0.05,k)
1	15.21	8.56	3.84
2	20.58	14.87	5.99
3	38.95	26.80	7.82
4	51.12	27.64	9.49
5	57.13	30.97	11.07
6	66.30	33.78	12.59
7	95.91	34.55	14.07
8	152.14	37.12	15.51
9	162.17	40.00	16.92
10	164.66	43.53	19.68

Note: All T × R² values are greater than 5%, indicating "significance".

AIC and SC values for the VARMA(1,0,1)-GARCH(1,1) combination are the smallest; hence, the VARMA(1,0,1)-GARCH(1,1) combination is considered the most appropriate. Values of the various estimated parameters for the most appropriate VARMA(1,0,1)-GARCH(1,1) model are listed in Table 11.

Simulation Results

Table 11 shows the resulting equations and relevant results of carrying out the VARMA(1,0,1)-GARCH(1,1) model simulation. It indicates that when the current SO₂ is produced, the SO₂ concentration can be used for estimating the current CO concentration because the b₀ statistic of 11.88 is greater than 1.96, indicating significance. Additionally, the

one time lag SO₂ concentration also influences the current CO concentration as evidenced by the b₁ statistic of 5.47 exceeding 1.96, to indicate significance. These observations can be explained based on the incomplete combustion of fuel that leads to emission of SO₂ from industries and vehicles. The incomplete combustion, which is much faster than photo-oxidation reactions, causes the current CO. Additionally, the one time lag SO₂ concentration also influences the current CO because SO₂ in the atmosphere is not involved in photochemical reactions; thus, it does not affect CO formation after its release into the atmosphere. Additionally, CO in the atmosphere is not easily dispersed so that its accumulated concentration is rather high. The CO itself is also affected by its own one time lag because the a₁ statistic of 25.68 is greater than 1.96, which indicates significance. However, the current CO concentration is not significantly influenced by the two times lag value (a₂ statistic of -1.38 being less than -1.96 in a positive number indicates a lack of significance.

Analyses of Impact Responses

The analyses results for the lagged variables are listed in Table 12. The fourth term lagged variables having the lowest AIC value are selected for carrying out the impact response analyses for the fuel factor. Fig. 3 displays the impact responses for these two air pollutants to one unit variation.

Fig. 3(A) shows the impact response for SO₂ to variations of CO; the current SO₂ shows immediate response to variations in current CO. This indicates that the production of CO is mainly caused by incomplete combustion of SO₂-containing fuel that leads to an increase of atmospheric CO concentration. This conclusion is similar to the simulated

Table 10. Results of the VARMA-GARCH examination for the various fuel factors.

VARMA	ARCH	ARCH(1)		ARCH(2)		GARCH(1,1)		GARCH(2,1)	
	AIC	SC	AIC	SC	AIC	SC	AIC	SC	
VARMA(1,0,0)	1.302	1.317	1.889	1.903	1.311	1.298	1.470	1.486	
VARMA(2,0,0)	1.260	1.277	1.216	1.230	1.202	1.231	1.582	1.599	
VARMA(0,0,1)	1.435	1.473	1.399	1.404	1.420	1.427	1.405	1.423	
VARMA(1,0,1)	1.222	1.238	1.180	1.199	1.159	1.178	1.179	1.192	
VARMA(2,0,1)	1.231	1.249	1.857	1.883	1.176	1.190	1.175	1.194	

Table 11. Parameters obtained using the combined vector model and GARCH(2,1) models for the various fuel factors.

	a ₀	a ₁	a ₂	b ₀	b ₁	b ₂	d ₁	α ₀	α ₁	α ₂	β ₁
VARMA(1,0,0)	0.88	0.84		1.08	0.004			0.05	4.76	0.02	3.23
t-statistic	17.1	1.56		2.01	-0.65			43.1	3.45	0.51	3.90
VARMA(2,0,0)	1.01	1.13	-0.28	0.003	1.24	5.67		0.04	3.41	0.04	0.96
t-statistic	5.46	6.89	-37.38	-1.12	0.98	-0.07		27.9	-5.23	4.73	-1.91
VARMA(0,0,1)	1.11			0.003			0.64	0.04	3.03	0.06	5.23
t-statistic	15.2			27.78			2.51	-2.64	1.54	-6.76	18.71
VARMA(1,0,1)	1.03	0.80	-0.11	0.16	0.32		0.36	-0.04	2.96	-3.56	2.89
t-statistic	2.67	25.68	-1.38	11.88	5.47		4.55	6.92	-1.28	-13.4	1.97
VARMA(2,0,1)	1.04	0.66	2.56	0.003	12.34	-2.41	0.04	0.04	2.98	0.04	-2.34
t-statistic	-2.16	30.12	1.06	1.90	5.56	-0.03	26.45	-0.09	3.56	-3.98	0.06

$$CO = a_0 + a_1CO_{(t-1)} + a_2CO_{(t-2)} + b_0SO_{2(t)} + b_1SO_{2(t-1)} + b_2SO_{2(t-2)} + d_1\varepsilon_{t-1}$$

$$h_t = \alpha_0 + \alpha_1 \varepsilon_{t-1}^2 + \alpha_2 \varepsilon_{t-2}^2 + \beta_1 h_{t-1}$$

Table 12. AIC values of the lagged variables for the various fuel factors.

Lag items	AIC
1	-5.89
2	-6.71
3	-8.02
4	-8.56*
5	-7.92
6	-8.14
7	-6.57
8	-7.00
9	-6.93
10	-6.88

results shown in previous paragraphs that indicated that when the current SO₂ is produced, the SO₂ concentration can be used to estimate the current CO concentration. Fig. 3(A) also reveals that CO is significantly affected by the current CO and the one time lag CO concentrations.

Fig. 3(B) shows the impact response for CO to variations in SO₂; the current CO shows a response when variations in SO₂ occur. This indicates that the incomplete combustion of SO₂ will lead to the production of current CO with lagging. The figure also shows SO₂ obviously responds to the current and one time lag concentrations so that when the current variation occurs, the current concentration responds

with a significant influence even on one time lag or two time lag concentrations. It can be seen in Table 5 that the b₁ statistics for the one time lag SO₂ of 2.56 is greater than 1.96.

CONCLUSIONS

The factor analyses carried out using the multivariate statistical analysis in this research leads to the selection of two relatively important factors, i.e. the photochemical pollution factor and the fuel factor because the air pollutants contained in these two factors cover the 5 air pollutants in the air pollution indices promulgated by the Environmental Protection Administration (Taiwan). Results obtained by using these two factors will therefore fully reflect the air quality conditions for the Central Taiwan Air Quality Total Quantity Control District.

The analysis results show that PM₁₀ has a low skewness of 0.86 because PM₁₀ is the major air pollutant in the Central Air Quality Total Quantity Control District. Hence, higher PM₁₀ concentration leads to more serious air pollution especially during the winter time when the atmospheric PM₁₀ increases. Additionally, the model simulation results reveal that the current O₃ concentration is influenced by the first time lag and the two time PM₁₀ concentrations as well as the one time lag NO₂ concentration. In other words, the current O₃ concentration is influenced by the previous PM₁₀ and

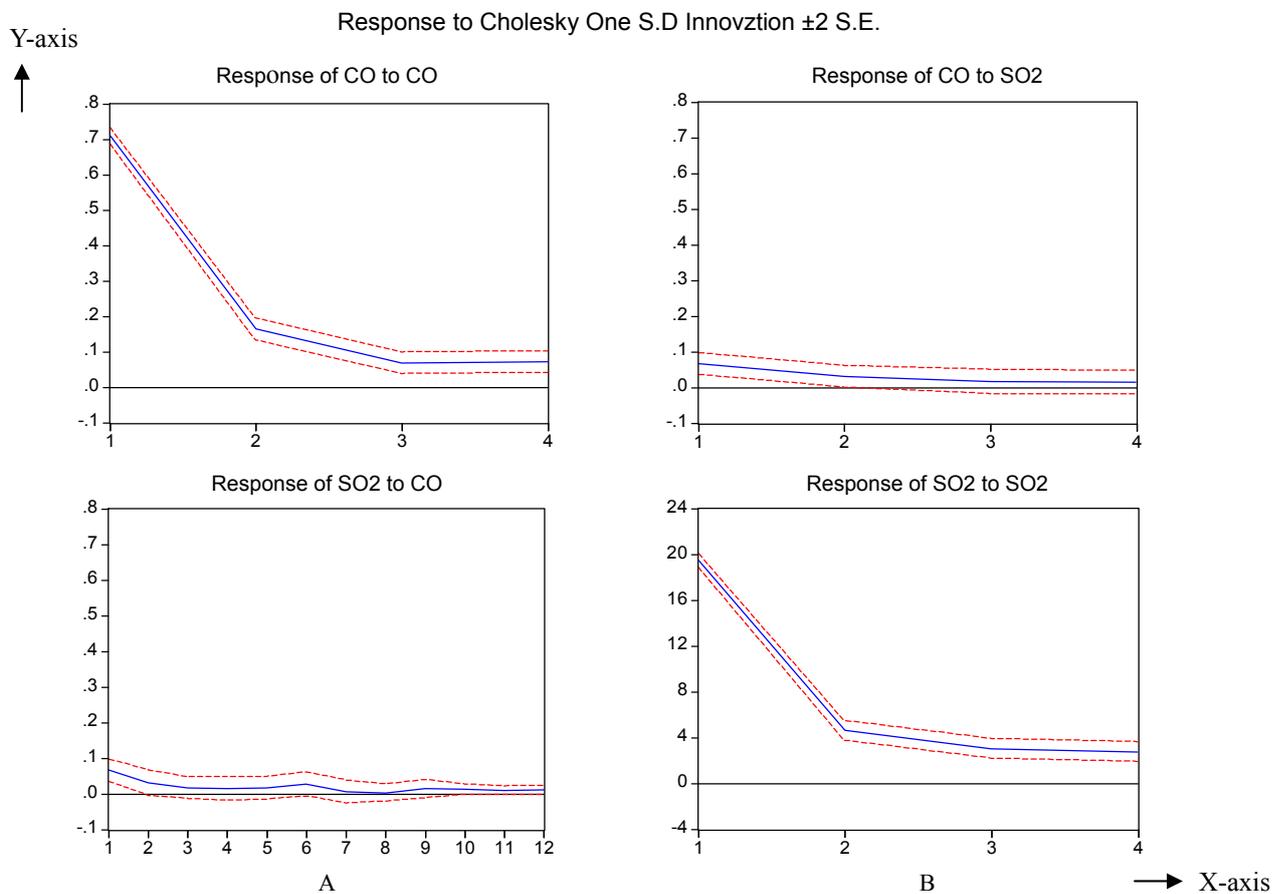


Fig. 3. Analyses of impact response for the various fuel factors.

NO₂ concentrations. When the current SO₂ is produced, the current CO concentration can be estimated based on the SO₂ concentration, and the current CO concentration is influenced by the one time lag SO₂ concentration.

Results of impact response analyses show that O₃ concentration does not respond to variations in current O₃ and NO₂ concentrations until after the one time lag. When PM₁₀ and NO₂ are produced, their concentrations cannot be used for predicting the immediate current O₃ concentration until the one time lag is reached. The SO₂ concentration responds to the variations in the current CO, indicating that the production of CO is caused by incomplete combustion of SO₂-containing fuel, thus increasing CO concentration. Responses of CO concentration to changes in current SO₂ concentration also indicate that during incomplete combustion, SO₂ leads to the production of current CO without lagging.

The integrated VARMA-GARCH model used in this research is capable of evaluating the degree of instantaneous variations of each air pollutant in the air quality total quantity control district. Its implementation is expected to improve the model simulation results significantly by considering the heteroscedastic characteristics of data series that have been ignored in previous research. In this paper, the vector time series is coupled with the (G)ARCH model to convert simple data series into valuable information so that raw data are better and more completely presented for the purpose of revealing future trends in variation.

As mentioned earlier, the classic regression analysis model assumes that variables of residual values are constants and that the expected values for every period remain the same. However, the results of many studies indicate the time-varying nature of sequence order variables. These observations have been confirmed in various fields such as economics, banking and financing, but not in environmental engineering applications. The authors are currently conducting empirical research intended to verify the subject in question with experimental data, and we will publish the results in the near future.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the support of the National Science Council of Taiwan under Grant NSC-2011-2221-E-214-011. The authors also deeply appreciate the editor and the anonymous reviewers for their insightful comments and suggestions.

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Received for review, November 6, 2011
Accepted, April 5, 2012