

Monitoring the Quality of a Chemical Production Process Using the Joint Estimation Method[†]

Sameer Prasad

Department of Management, School of Business Administration, The University of Texas—Pan American,
1201 West University Drive, Edinburg, Texas 78539-2999

David Booth^{*‡} and Michael Y. Hu[§]

Department of Administrative Sciences and Department of Marketing, Graduate School of Management,
Kent State University, Kent, Ohio 44242

Received May 20, 1994[®]

The standard approach to monitoring a chemical production process is the control chart. Such a chart assumes that the data values are independent and identically distributed. It has been shown that such is not so for many production processes especially those of chemical interest. However a more realistic approach to the problem can be developed using time series modeling. A recent development in time series methodology, the joint estimation procedure, allows for the detection and identification of four types of outliers. This study draws the correspondence between these outlier types and out-of-control situations (that is, adverse process changes) in chemical process control. The correspondence is illustrated in this study using a dye liquor data set. Results show that the joint estimation procedure is appropriate for use in process control and provides advantages for dealing with out-of-control problems in production processes.

1. INTRODUCTION

Control charts, such as the Shewhart and CUSUM charts, are used extensively in the control of production processes. An aberrant observation, (outlier) is interpreted as an out-of-control point. When an outlier is observed, the search begins for an assignable cause, i.e., the physical reason for the process being out-of-control. Once the cause is found, it is corrected and the production process goes on.

To use standard control charts several statistical assumptions must be met.¹⁴ One of the most important is that the random variables describing the underlying production process must be independent and identically distributed (iid). The assumption of independent and identically distributed random variables is often violated.^{2,3} In many industrial applications autocorrelations among variable values are found. It has been shown that the violation of the iid assumption will lead to inaccurate specification of α and β (the probability of type I and II errors, respectively) and consequently render the control chart methods unreliable.

In this paper, we will show how robust (outlier resistant) time series methods can be used to correct the problems associated with the violation of statistical assumptions and further how these methods also provide identification of various types of outliers. The identification of outlier type will be shown to provide valuable information for the analyst. Furthermore, the robust time series methods will be shown to be superior in yielding more timely detection of outliers. The performance of a recently developed robust procedure, joint estimation, will be compared to a previously suggested approach, the M-type iterative method. The comparison will be illustrated using an actual chemical production data set.

First, we will discuss the problems associated with the control chart methodology. Next, we review the use of time series methods for statistical process control (SPC) together with a discussion of this new procedure. We then illustrate the application of the joint estimation procedure using the dye liquor data.¹⁴ Then, finally, we will discuss implementation issues arising from the use of the joint estimation procedure.

2. PROBLEMS WITH STANDARD CONTROL CHARTS

Common problems regarding the use of standard control charts include (1) having too many false alarms, (2) the inability to detect process problems, and (3) delayed detection of out-of-control points. It is important to remember that false alarms can be costly. They often result in shutting down a production process or possibly discarding part of the production lot. Such unnecessary stoppages reduce overall productivity. The inability to detect quality problems can lead to lower consumer satisfaction and an overall decline in the goodwill of the firm. Finally, if out-of-control situations are detected too late, one may incur substantial losses.

False alarms and the inability to detect problems are inversely related. We adopt the model and notation due to Speed and Culpin²⁰ for the quantification of the trade off between the costs of false alarms and the inability to detect quality problems.

Let

f = the cost of a false alarm,

d = the cost of not detecting a quality problem,

α = the probability of a false alarm,

δ = the probability of discovering a process problem,

π = the probability of a process problem,

and $E(\alpha)$ = the expected cost.

Then $E(\alpha) = \pi(1 - \delta)d + (1 - \pi)\alpha f$

[†] The authors would like to thank Chung Chen for allowing the use of the SCA program.

^{*} Department of Administrative Sciences.

[‡] Department of Marketing.

[®] Abstract published in *Advance ACS Abstracts*, December 15, 1994.

where $\pi(1 - \delta)d$ is the overall expected cost of not detecting a quality problem and $(1 - \pi)\alpha f$ is the overall expected cost for false alarms.

The objective here is to minimize the expected cost $E(\alpha)$, by using an optimum $\alpha = \alpha_0$, such that the overall expected cost of not detecting quality problems and $(1 - \pi)\alpha f$, the overall expected cost for false alarms, are equal. Thus process control can be achieved by setting up control bands as a function of the critical value (based on α_0) and the variability in the data. It should be noted that the specification for the cost of errors is pertinent to any outlier detection procedure, control chart, or time series based methods.

In industrial applications, two additional issues have to be addressed—improper training of the quality control operators and the use of statistical procedures that are not powerful enough to detect out-of-control points. Improper operator training leads to the inappropriate selection of α_0 , or selecting control limits that do not reflect the specified α_0 . Errors in calculating the standard deviation and central tendency also can distort the control limits. More problematic would be the use of low power $(1 - \beta)$, i.e., insensitive statistical procedures to detect out-of-control situations. This may be due to the assumptions carried with the control chart: the observations in the process are independent of each other, and the outliers do not affect the estimation of the process mean and standard deviation. Next we review the development of robust time series methods, which have been applied to deal with the limitations of the Shewhart chart.

3. ROBUST TIME SERIES METHODS AND OUTLIERS

Now we will discuss the application of time series analysis to SPC. We first look at how time series methods are used to account for autocorrelation in a production process. Following that, we examine the way in which outliers are determined from the series and thus can be used to locate process quality problems. Finally, we explore the distortions due to outliers on the estimation of the model parameters as well as the identification of out-of-control situations.

3.1. Time Series for Statistical Process Control. In the early 1970s, it became apparent that it was very important to manage inventories of weapons grade nuclear material and similar fuel material used at power plants, e.g., special nuclear materials (SNM). The accounting systems used in controlling these inventories are based on material balance accounting.^{8,13} It was essential that statistical methods for detecting losses from SNM inventories be as close to real-time as possible. Because the material balance accounting system was based on sequential measurements of SNM in a particular material balance area, it was natural to deal with the problem of identifying losses by using standard control chart methods. As Speed and Culpin²⁰ point out, the use of control charts without any special theory and without taking into account of the correlation between successive measurements has been unsuccessful for SNM. To remedy these problems, Downing et al.¹¹ proposed modeling the process data using the Box-Jenkins¹⁷ (ARMA) time series approach. Let Z_t be a time series following a general ARMA process as¹⁷

$$\phi(B)Z_t = \theta(B)a_t, \quad t = 1, 2, \dots, n$$

where n is the number of observations for the series, B is

the back shift operator such that⁷

$$\phi(B) = 1 - \phi_q B - \dots - \phi_p B^p$$

and

$$\theta(B) = 1 - \theta_1 B - \dots - \theta_q B^q$$

are two polynomials in B with orders p and q , and a_t 's are independently and identically distributed with mean zero and constant variance σ_a^2 .

More recently, Alwan and Roberts² proposed another approach to SPC using ARMA models. They also attacked the problem of correlation between successive observations that standard control chart methods ignored. With the ARMA model, they developed two charts, the common-cause chart and the special-cause chart, to detect process difficulties. Although, such procedures² build in the correlation among observations, they are unable to cope with outliers that affect the estimates¹⁵ of the central tendency, variance, and the correlation of the underlying series being monitored. Outliers are large errors which are not modeled by the underlying specified ARMA series. Consequently, improper estimates distort the monitoring process, which would lead to inappropriate levels of false alarms and the power of the test. Next we will examine how robust times series methods have been applied to obtain reliable monitoring systems.

3.2. Outliers and Robustness. To resolve the problems caused by statistical outliers, Denby and Martin⁹ developed Generalized M-estimation (GM), a robust (outlier resistant) method to better estimate the AR(1) series parameter in the presence of time series outliers. In addition, Martin and Zeh¹⁶ developed graphical techniques to identify two types of time series outliers in the AR(1) case. Further, Denby and Martin⁹ developed the modern terminology for Fox's¹² two types of outlier. An outlier is defined as an underlying stochastic process that generates an outlying observation.⁹ Denby and Martin called additive outliers (AOs) those from outlier processes that affect only a single observation, while an innovational outlier (IO) is from an outlier process "in which a single innovation is extreme". Note that here "innovation" implies a change in the underlying stochastic process, and thus an IO will affect subsequent observations while an AO will not. These outliers can then be directly related to a production process.

Outliers are the out-of-control points in a production process. Booth³ pointed out that AOs signal a one-time process problem (e.g., a one-time bad batch in a production process). Further, an IO signals a continuing process problem (e.g., a continuous deterioration in a production process caused by wear in a machine, etc.). Booth³ and Booth et al.⁵ give practical examples of the use of these methods. In particular, they showed that knowing outlier type helps in determining assignable causes. In addition, Acar and Booth¹ discussed the implementation of such methods for real-time SPC. However, monitoring with the GM is limited to only processes that can be defined in terms of AR(1).

Prasad¹⁸ was able to generalize robust time series methodology to the entire range of ARMA(p,q) processes. Using the M-type iterative procedure proposed by Chang et al.,⁶ Prasad¹⁸ showed that it was possible to develop a general ARIMA model for a production process that detects outliers

and types the outliers as AO or IO. Assuming only one outlier occurred in the series at $t = t_1$ and $I_i(t_1) = 1$ when $t = t_1$, $I_i(t_1) = 0$ otherwise, the observed series $\{Y_i\}$ can be expressed as follows:⁷

$$Y_i = Z_i + \omega L(B)I_i(t_1)$$

where ω and $L(B)$ denote magnitude and the dynamic pattern of the outlier effect. The outlier types are defined as

$$L(B) = \frac{\theta(B)}{\phi(B)} \quad \text{for IO}$$

$$L(B) = 1 \quad \text{for AO}$$

Although the M-type iterative procedure can be relatively robust, the procedure is cumbersome for practical applications. Outliers are located by the initial nonrobust time series estimates, and it is then necessary to specify intervention models to accommodate their influence. Consequently, there is a tendency for masking (unable to detect process changes) and spurious reading (false alarms). Given this, the M-type iterative procedure performs less than adequately.

More recently, Chen and Liu⁷ developed a new procedure that reduces the possibility of masking and spurious observations by jointly estimating the series and locating outliers. This procedure is termed the joint estimation procedure. Here, it is not necessary to specify intervention model to obtain robust estimates. In addition the procedure can detect not only the initial two types of outliers but also four. The additional outlier types are termed level shift (LS) and temporary change (TC). One may consider TC to range between the extremes of a one-time change (AO) and a permanent shift in the level of the process (LS). The two additional outliers are defined as⁷

$$L(B) = \frac{1}{(1 - B)} \quad \text{for LS}$$

and

$$L(B) = \frac{1}{(1 - \delta B)} \quad \text{for TC } (0 < \delta < 1)$$

The parameter δ affects the rate at which aberrations are dampened. In the case of AO, δ would be 0 and for the LS δ would be 1. For the TC it is recommended⁷ that δ to be set at 0.7.

As we will see later, these outlier types will help to determine assignable causes of variation. Next we will describe the joint estimation procedure and its implications for SPC. Following this description we will provide an example where the benefits of using this new procedure for SPC applications becomes evident.

4. METHODOLOGY

4.1. The Joint Estimation Procedure. In this paper, the joint estimation procedure recently developed by Chen and Liu⁷ will be used to model a chemical production time series. This new method is superior to the one used by Prasad,¹⁸ in that (a) outliers are obtained iteratively based on the adjusted residuals and observations; (b) the procedure does not require intervention models to be estimated to accommodate the outliers; (c) the identification and location of outliers are

based on robust parameter estimates, thus the model parameters are more reflective of the representative data; and (d) the outlier effects are jointly estimated using multiple regression.⁷ Robust parameter estimates are computed using the regression results to accommodate the outlier effects. Thus, the procedure differentiates and accommodates four forms of aberrations. Unlike transfer function modeling, for example, dynamic regression,^{17,21} there is no prior need to assume the location and type of problem.

The joint estimation procedure consists of a three-stage process: first, it provides the initial parameter estimates and conducts potential outlier identification; second, it conducts joint estimation of outlier effects, and estimates of model parameters are obtained using the information from the previous stage; and third, the identification of the actual outliers and their effects based on the robust (outlier resistant) parameter estimates are found.⁷ Full details are available in the references. The computer program implementation is available from Scientific Computing Associates (SCA) Corporation, Chicago, IL.

4.2. Joint Estimation Procedure for SPC Applications.

The joint estimation procedure provides information on changes in a chemical process by looking at the location and identification of outliers. The procedure differentiates and accommodates four different forms of aberrations: innovational outliers (IO), additive outliers (AO), level shifts (LS), and temporary changes (TC).⁷ An AO could possibly signal a one-time problem; a LS would indicate a continuing change in the remaining part of the series. Operators should use this information to explore the causes for such departures. These aberrations can be tied to various forms of quality and measurement problems: for example, as a one-time problem (additive outlier); a continuing problem in the remaining part of the modeled production process (level shift); and a range of intermediary effects that can be picked up as innovational outliers and temporary changes. In addition, in this procedure the parameter estimates are robust to the influence of outliers. In process control applications we can expect the actual probability of false alarms (α) to be close to the desired (set) α_0 , and thus to at least approximately minimize the overall cost in running the monitoring system. In operational terms this translates to preventing excessive false alarms or masking actual quality problems.

As a demonstration of this new procedure for statistical process control we will apply it to a data set previously reported in the literature and compare the results to those from using the Shewhart control chart and the previously proposed M-type iterative method. The joint estimation procedure will be judged in terms of the accuracy of modeling the underlying process and the ability to signal various types of quality problems.

5. ILLUSTRATION OF THE JOINT ESTIMATION PROCEDURE FOR SPC

To illustrate the capabilities of the joint estimation procedure for SPC applications we will apply it to the monitoring of acidity in dye liquor. In this section, we will describe the data set and the respective runs of the various monitoring procedures. We will then discuss the implications of the joint estimation procedure for SPC applications.

Table 1. Parameter Estimates and Out-of-Control Points

		Shewhart chart	M-type iterative procedure	joint estimation procedures
parameter	C or mean (<i>t</i> -value)	4.2626	1.2877 (2.81)	1.0717 (2.44)
estimates	AR(1) (<i>t</i> -value)	NA	0.6962 (6.51)	0.7476(7.22)
	residual error	0.1686	0.0925	0.0790
outliers	observation, type	35	35 AO -0.43 (-5.07)	09 IO 0.231(2.93)
	estimate (<i>t</i> -value)			16 AO 0.213(3.37)
				35 AO -4.29(-6.78)
specified test level	critical value (α_0)	2.50 (0.0125)	2.50 (0.125)	2.50 (0.1254)

5.1. Data. The data set is taken from Grant and Leavenworth¹⁴ (p 93). They describe the data set in the following manner.

In the dyeing of woolen yarns, it is desirable to control the acidity of the dye liquor. Unless the dye liquor is sufficiently acid, the penetration of color is unsatisfactory; on the other hand, a too-acid liquor affects the durability of the products made from the yarn. Acidity is conveniently measured as pH (the negative common logarithm of the hydrogen ion concentration). A low pH corresponds to high acidity and vice versa. In any dyeing operation there is a band of pH values within which the best results for which color penetration and durability are obtained.

Average pH values across five Hussong kettles over time are given in the data. The authors detail the following assignable causes of variation.

"The acidity of the dyeing solution depends not only on the constituents put into the dye liquor but also on the characteristics of the wool being dyed. From time to time it is necessary to use wools from sources that have different characteristics. Although blends of wools from various sources are made, successive blends will differ somewhat from one another.

On February 1 a new blend of entirely different wools was introduced. Immediately the acidity dropped. On February 5 (observation 9) after the old surplus stock had been used up, acidity fell until corrective measures were taken on February 8 (observation 16). At this time the amount of acid introduced into the dye liquor was changed.

Thereafter, with the exception of a brief departure from control on February 21 (observation 35), the pH values continued in satisfactory control. The temporary difficulty on February 21 was traced to two batches of improperly neutralized carbonized (baked with concentrated sulfuric acid) stock. Such stock is acid in relation to stock normally used."

5.2. Procedure Comparisons. Three procedures were utilized to locate quality problems in the pH dye liquor data set: Shewhart control chart,¹⁴ M-type iterative, and the joint estimation procedure. The time series methods (M-type iterative and the joint estimation procedures) will be compared to a procedure that does not build in the correlation among successive observations (Shewhart chart). In addition, we will compare the joint estimation procedure with the M-type iterative procedure to show impact to modeling additional forms of outliers and the benefits of jointly estimating outliers and parameter estimates. To establish a common basis for comparison, a critical value of 2.50 or $\alpha_0 = 0.0125$ is specified for all three procedures. The three procedures will be compared in terms of the number of outliers detected and false alarms.

For the two time series methods it was first necessary to identify the type of ARMA(*p,q*) model that would be the

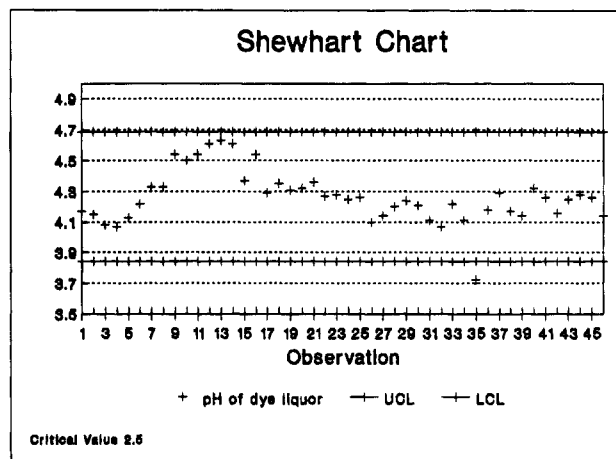


Figure 1. The detection of quality problems in the pH level of dye liquor using the Shewhart chart.

most appropriate. By looking at plots of the autocorrelation and partial autocorrelation, we ascertained that the underlying process could be best represented by an AR(1) model. The software package by SCA was used to obtain outliers and parameter estimates for both the M-type iterative procedure and the joint estimation procedure. For the Shewhart chart, we computed the standard deviation and mean from the original series $\{Y_t\}$, to set up the confidence interval (control bands) accordingly. Observations exceeding the control bands were identified as out-of-control points.

5.2.1. Modeling the Underlying Process. In Table 1, we notice that the residual error (or the unexplained variance) falls as we go from the Shewhart chart (nontime series approach) to time series methods. Comparing the two time series methods, a further reduction in the residual error is observed when the joint estimation procedure is used. In addition, the AR(1) coefficient becomes more statistically significant as evidenced by the larger *t*-values.

5.2.2. Out-of-Control Situations. In Figure 1, we notice that the Shewhart chart locates only one observation (35) as out-of-control, and this reflects the temporary difficulty of the improperly neutralized carbonized stock. The M-type iterative procedure in Figure 2 also locates an out-of-control situation at observation 35, but signals it as an AO. This reflects a temporary problem on February 21 (observation 35). We observe the joint estimation procedure, as shown in Figure 3, not only locates the aberration at observation 35 but also locates two other outliers. An IO is picked up at observation 9 and indicates a continuing problem. Upon looking at the documentation we find that on February 5 (observation 9) there was a continuous change in acidity due to a gradual change in the blend of wool. However, on February 8 (observation 16) a corrective action took place, and the procedure picked it up as an AO. This indicates that there was only a one time change in the process and

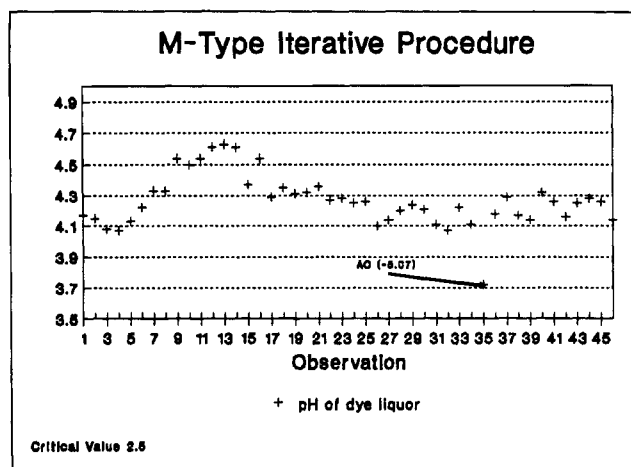


Figure 2. The detection of quality problems in the pH level of dye liquor using the M-type iterative procedure.

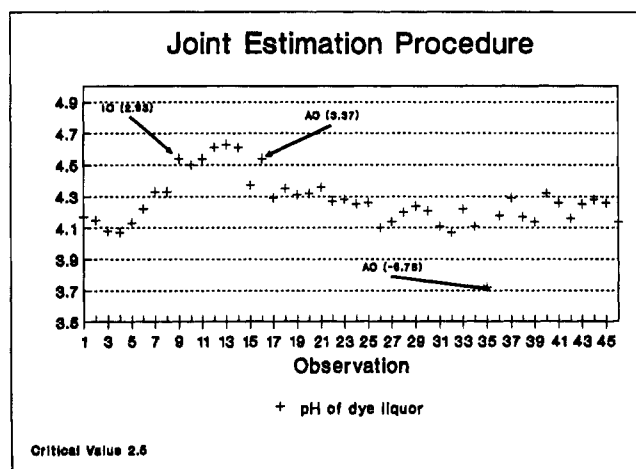


Figure 3. The detection of quality problems in the pH level of dye liquor using the joint estimation procedure.

needs to be explored further. This was somewhat unexpected in that a continuing change was expected and should be investigated further. In this example we found the joint estimation procedure to be more useful in that it provides more information in monitoring and modifying the process.

5.2.3. Discussion. In our comparisons we notice the benefits of using time series methodology and the importance of having robust parameter estimates. We can exploit the correlation structure of the time series methods to flag the type of quality problems. In comparing the joint estimation procedure with the M-type iterative procedure we notice the benefits of having robust parameter estimates. In this example, we show the joint estimation procedure for the specified α has the most power ($1 - \beta$), because in Table 1 for the specified α we are able to locate more of the actual quality problems (outliers). This is due to the fact the parameter estimates are more robust, and consequently the actual α is closer to the desired optimum α_0 .

There is another important point. If the joint estimation procedure is to be an on-line real-time method for SPC it must determine whether an observation is an outlier when the measurement is made. More retrospective methods are of lesser use to practitioners. In the original literature,^{7,18} the discussion shows that is exactly what happens with the methods discussed herein. Thus the methods considered are reasonable to use in practice. One potential problem of joint estimation is that it cannot identify an outlier that is an

immediate point. It must wait one period for that.⁷ This is a slight problem because the analyst immediately knows there is a problem but must get another observation to get information on problem type. If problem type information is not desired, then the result is immediate. Although this is a slight drawback, it is an improvement over the standard control chart, which can require as many as seven extra observations to make an out-of-control determination.^{2, 23, 24}

6. ADVANTAGES OF THE JOINT ESTIMATION PROCEDURE

The joint estimation procedure has a number of advantages over other methods. These advantages include

(1)The out-of-control observations are easily detected especially when the latest observation is such.

(2)The sensitivity of the method can be adjusted (by adjusting the desired probability of a type I error) to any appropriate level.

(3)The method is useful for many processes, especially those of a chemical nature. For example, it can be used for detecting losses (e.g., thefts) from a nuclear material inventory, a pharmaceutical-inventory, or any other inventory based on a material balance accounting system.⁴

(4)The procedure can deal with any type of time series model. Other methods are often more specialized, e.g., the GM 4 approach.

(5)The procedure identifies all major outlier types, thus giving more information on possible assignable causes than other available methods.

(6)The approach using standard statistical methods⁴ can provide estimates of the amount of material lost from an inventory¹³ that other methods often do not supply.

(7)The procedure provides more information per computer run than do methods based on deleted observations.⁴

These advantages make the joint estimation approach very attractive though there is the drawback of the amount of training required by the operator, as discussed in the following section.

7. IMPLEMENTATION

Implementation issues for time series methods have been discussed by Alwan and Roberts² and Acar and Booth.¹ Following those thoughts, in this section we examine the application of real-time process control with the joint estimation procedure. In implementing the joint estimation procedure it will be necessary to have the appropriate hardware, software, and support personnel. Unlike the control chart which can be set up by a technician with a calculator, the joint estimation procedure requires a person who is familiar with time series methodology to identify the type of ARMA(p, q) series exhibited by the chemical production process. In addition, the cost to run the appropriate software (hardware and software) would run less than \$1000 per year.

Difficulties that are expected to arise in implementing the joint estimation procedure revolve around the level of training required for operators to identify time series models. To simplify the process of model selection, expert systems are available commercially (e.g., SCA) that provide model identification based on the data provided. In addition, we have some indications¹⁹ that the joint estimation procedure is relatively robust to model misspecification, which further

eases the implementation process. Although the cost to implement the more sophisticated joint estimation procedure is greater than that of the Shewhart chart, the savings incurred due to fewer false alarms and the ability to locate and identify outlier types and process problems should more than compensate for these additional costs.

The additional information provided by the outlier identification can be used to signal the type of change required to adjust the process back into a state of control. This feedback control loop can be automated, reducing the time that the process is out-of-control. Coupled with the fact that time series methods pick up problems earlier,¹⁰ it may be possible to achieve automatic real-time process control¹.

8. CONCLUSIONS

On the basis of the results of this study it can be concluded that (1) the joint estimation procedure is a successful method for identifying outlier types (and thus a help in discovering assignable causes of variation) in production processes; (2) the procedure is a useful method for production process control and chemical inventory management; and (3) the procedure provides clear advantages over the other available methods. Based on these results, we recommend joint estimation as a useful addition to the control of chemical production processes.

REFERENCES AND NOTES

- (1) Acar, W.; Booth, D. E. Easier Quality Control: Combining Problem Classification With Time Series Analysis. *Production Inventory Mgmt* **1987**, 28, 53–58.
- (2) Alwan, L. C.; Roberts, H. V. Time Series Modeling for Statistical Process Control. *J. Business Economic Statistics* **1988**, 6(1), 87–95.
- (3) Booth, D. E. *Some Applications of Robust Statistical Methods to Analytical Chemistry*. The University of North Carolina at Chapel Hill, Unpublished Ph.D. Dissertation.
- (4) Booth, D. E. A Method for the Early Identification of Loss From a Nuclear Materials Inventory. *J. Chem. Inf. Comput. Sci.* **1986**, 26, 23–28.
- (5) Booth, D. E.; Acar, E.; Isenhour, T.; Akham, S. Robust Time Series Models and Statistical Process Control. *Industrial Mathematics* **1990**, 40 (part 1), 73–97.
- (6) Chang, I.; Tiao, G. C.; Chen, C. Estimation of Time Series Parameters in the Presence of Outliers. *Technometrics* **1988**, 30(2), 193–204.
- (7) Chen, C.; Liu, L. Joint Estimation of Model Parameters and Outlier Effects in Time Series. *J. Am. Statistical Assoc.* **1993**, 88, 284–298.
- (8) Chernick, M. R.; Downing, D. J.; Pike, D. H. Detecting Outliers in Time Series Data. *J. Am. Statistical Assoc.* **1982**, 77, 743–747.
- (9) Denby, L.; Martin, R. D. Robust Estimation of the First Order Autoregressive Parameter. *J. Am. Statistical Assoc.* **1979**, 74, 140–146.
- (10) Domangue, R.; Patch, S. C. Some Omnibus Exponentially Weighted Moving Average Statistical Process Monitoring Schemes. *Technometrics* **1991**, 33(2), 299–313.
- (11) Downing, D. J.; Pike, D. H.; Morrison, G. W. Analysis of MUF Data using ARMA Models. *Nuclear Materials Mgmt.* **1978**, 7(4), 80–86.
- (12) Fox, A. J. Outliers in Time Series. *J. Royal Statistical Soc. Ser. B* **1972**, 34, 340–363.
- (13) Goldman, A. S.; Picard, R. R.; and Shipley, J. P. Statistical Methods for Nuclear Material Safeguards: An Overview. *Technometrics* **1982**, 24, 267–274.
- (14) Grant, E. L.; Leavenworth, R. S. *Statistical Quality Control*, 5th ed.; New York, McGraw-Hill Book Co.: 1980.
- (15) Hogg, R. V. Statistical Robustness: One View of its Use in Applications Today. *Am. Statistician* **1979**, 33, 108–115.
- (16) Martin, R. D.; Zeh, J. E. Determining the Characteristics of Time series Outliers. *Proceedings of the American Statistical Association; American Statistical Association: Business and Economics Section*, Washington, DC; 1977.
- (17) Pankratz, A. *Forecasting with Dynamic Regression Models*; New York: Wiley-Interscience, 1991.
- (18) Prasad, S. *A Robust Monitoring Method for the Early Detection of Deviations in a Time Series Process with Applications in Operations Management*. Kent State University, Kent, OH, Unpublished Ph.D. Dissertation.
- (19) Prasad, S.; Booth, D.; Hu, M.; Deligonul, S. The Detection of Nuclear Material Losses by Means joint estimation of Parameters and Outlier Effects in Time Series. Submitted for review to *Decision Sci.*
- (20) Speed, T. P.; Culpin, D. The Role of Statistics in Nuclear Materials Accounting: Issues and Problems. *J. Royal Statistical Society, A* **1988**, 149, Part 4, 281–313.
- (21) West, M.; Harrison, P. G. Monitoring and Adaptation in Bayesian Forecasting Models. *J. Am. Statistical Assoc.* **1986**, 81, 741–750.

CI940060M