Using Polynomial Smoothing and Data Bounding for the Detection of Nuclear Material Diversions and Losses

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Successive nuclear material balances form a time series of often autocorrelated observations. Significiant deviations from the underlying in-control process model or time series pattern, i.e., outliers, indicate an adverse process change or out-of-control situation relative to the model. This paper uses these ideas to demonstrate a method for detecting nuclear material losses earlier than currently used models and thus helps to alleviate a problem which is growing to world crisis proportions. The process control capabilities of these methods were successfully tested on two autocorrelated data sets of nuclear material balances with known removals. These algorithms should be of assistance to the nuclear engineer involved in process control.

A. INTRODUCTION

In Nuclear Materials Accounting the goal is to accurately discover material losses (e.g., leakage or theft) as early as possible to minimize the threat to life and property. Despite some progress, control against leakage, theft, or loss of radioactive material has continued to be a problem. International terrorist activity and the desire of renegade nations to become nuclear powers make the threat of theft very real. Only recently it was discovered that controls for weapon grade nuclear materials are grossly deficient in the former Soviet Union as evidenced by several instances of stolen plutonium and uranium being found in Germany.[1] Possible scenarios regarding the theft of weapons grade material are frightening.

In addition, leakage of radioactive material into the environment can similarly affect the well being of millions of people. Thus any method that could detect such losses more quickly and accurately would be indeed a significant contribution. This is very important whether the losses are continuous or sporadic, including small quantities over a period of time.

We show in this paper that nuclear material loss situations may be recognized through the detection of statistical outliers.[38,44,45] In general terms, an outlier is an inconsistent or nonrepresentative point in a data set that causes a difficulty or problem in a statistical analysis. Traditional statistical process control (SPC) charts as well as most of the other methods in current use are based upon the assumption that the system observations in the time series are independent[43] and identically distributed (IID) about the targeted process mean (u_0 or TV) at any time t and that the distribution is normal when the process is in statistical control.[2] Independence implies that there is no particular pattern in the data.

Points outside of three standard deviations of the targeted process mean (u_0 or TV) are usually considered to be outliers. If such exist, the process is said to be "out of control". In other words, there is a significant adverse process change due to an assignable cause. Otherwise the process is said to be "in control", i.e., random variations of output within certain control limits.

Controlling the Process by Utilizing Time Series Models. In reality, the IID assumption of the conventional control chart often does not hold. In-control production process measures over time are often interdependent, i.e., the points are autocorrelated.[3–5] Under such conditions, traditional SPC procedures may be ineffective and inappropriate for monitoring and controlling product quality[2]—erroneously indicating an out-of-control situation when the supplementary criteria of the traditional control chart are applied. As we shall see, this is also true in nuclear material safeguards.

Many production process time series exhibit a characteristically repetitive pattern which can be mathematically modeled by an autoregressive moving average [ARMA(p,q)] model.[6] For example, ARMA(1,1) and other time series models have been empirically found in some cases to be appropriate for modeling a process time series.[7,2,8] Then an outlier, any point that deviates significantly from the underlying process model or time series pattern, indicates an out-of-control situation with respect to the process model. Such a point can be identified using statistical methods.

B. APPLICATIONS TO NUCLEAR MATERIAL SAFEGUARDS AND OTHER PRIOR RESEARCH

Statistical Process Control methodology is also applicable to other process industries such as the control of inventories of materials which are particularly valuable or involve critical safety concerns. Nuclear material safeguards are methods based upon nuclear material accounting to detect and prevent losses.

Marshall[9] criticized the International Atomic Energy Agency (IAEA) and the Nuclear Regulatory Commission (NRC) for weak safeguards regarding theft or loss of Special Nuclear Material (SNM), such as plutonium, uranium, and other radioactive substances. Despite some progress, control against leakage, theft, or loss of radioactive material has continued to be a problem.

Common statistical problems in material balance accounting are treated by Goldman et al.[10] in detail. These

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include additive errors that often occur in sums of material inventory measurements and transfers which usually are estimated. Estimates of material held up in equipment (in-process) are especially problematic. Speed and Culpin[11] note that it is extremely difficult to differentiate between a systematic error over several periods such as a poorly calibrated instrument (a discrepancy) and an outright diversion or theft. Bowen and Bennett[40] added to these reviews.

Statistical methods advocated or actually used today include the traditional Shewhart control chart, the cumulative summation (CUSUM) chart for detecting significant shifts in the mean or protracted losses, the sequential probability ratio test (SPRT), the power-one test statistic using material unaccounted for residuals (MUF_R), variations of the page test, the robust test, etc. Goldman et al.[10] and Speed and Culpin[11] found problems with all of these methods.

Speed and Culpin[11] further noted that none of the methods consider the prior probabilities of a diversion and of discovering the diversion nor the costs of a false alarm vs the cost of an undiscovered diversion. These are important for setting the control limits and thus the desired sensitivity of the methods. We note that Picard[12] concluded that the CUSUM test would be rather insensitive (low probability of detection) to protracted losses of material form large inventories.

The Material Balance Equation. The nuclear material balance at time t is defined as[34]

$$MB_t = I_t - I_{t-1} - T \tag{1}$$

where MB_t = observed material balance at time t (loss = $-$); I_t = inventory or quantity in the specified area at time t, including stock and material in-process; and T = net transfer accounted for (in = +, out = $-$) during the time interval. If T is positive, in-process and stock increase.

If there are no losses between time $t - 1$ and t, then $MB_t = 0$ and $I_t - I_{t-1} = T$, i.e., the net material transferred in (+) or out ($-$) to account for the change in inventory. Another term, often used in the nuclear industry instead of material balance, is material unaccounted for (MUF_t). To use MUF_t, both sides of the above equation are multiplied by -1 and $-MB = MUF$, being thus positive for a loss.[11] Equation 1 then becomes

$$MUF_t = I_{t-1} - I_t + T \tag{2}$$

or

$$MUF_{t+1} = I_t - I_{t+1} + T$$

A raw material inventory (of quantity I_t) consists of stock and the amount in-process. Transfer material (T) consists of new stock (+) and the raw material converted to waste and/or the final product leaving the process ($-$). It can be seen in eq 2 that if I_t is underestimated, $MUF_t > 0$. However, in the next period, $MUF_{t+1} < 0$ if I_{t+1} is estimated correctly.[11] On the other hand, if there is an actual loss at time t only and no measurement error, then $MUF_t > 0$. But $MUF_{t+1} = 0$ since all subsequent inventories will reflect that one time loss.

A negative material balance (positive MUF_t) would be due to such causes as leakage, unanticipated loss in effluents (waste), theft, accounting errors, systematic measurement errors such as poor instrument calibration, plugged probes, solid buildup in the process (e.g., settling in tanks), operator errors as misreading, mistranscribing, miscalculating, etc. What remains after all possible explanations are exhausted is the usual random error.[11]

A positive MB_t (negative MUF_t), an apparent gain, may be due to systematic measurement errors or an unaccounted for abnormal hidden amount in-process that later reversed itself as for example, caking on tube walls being released in some manner.

Since many estimates of the material are made in many parts of the process and there are numerous sources of error, often cumulative, seldom is $MUF_t = MB_t = 0$, the target value. Then a crucial question arises. Is a negative material balance due to normal statistical variations, a large error, or has there been a loss or diversion of nuclear material?

Material Balances As a Time Series. Even if no losses or gross errors have occurred, there will be variations among successive material balance or MUF measurements about the process mean, which is expected to be equal to 0. A succession of such material balance measurements forms a time series.

As with the control of a product specification in a production process, the output may be monitored for outliers which are defined as significant deviations from the process model or time series pattern in a direction away from the target value. For example, Downing et al.[13] applied ARIMA time series models to nuclear material balance data and developed a method to detect a case of constant loss. Pike et al.[14] and others also used time series methodology in the estimation of losses by subtracting the actual observation from the forecast based upon the model. Chernick et al.[15] used the **influence function** to detect a loss immediately after making an MB estimate. These authors noted that earlier methods required several inventory periods to detect a loss.

If a material balance is taken across a major part of a large process, hundreds of measurements and estimates may be necessary. The accumulation of many of these and other sources of error make it difficult to measure the dependency among observations. The parameters can only be estimated from the data, but outliers distort these estimates and inflate the standard deviation, thus resulting in erroneous conclusions. The methods that follow in this section are robust to the influence of outliers.

Booth[3,16] used the **generalized M estimator (GM) procedure**[41] to detect outliers in stationary nuclear material balance time series that fit an AR(1) model. Prasad[7] applied the M-type iterative procedure,[17] which is more general since it can be applied to all ARIMA models. Both methods also identify the type of time series outlier, i.e., additive or innovative which Fox[18] first identified.

Booth[3,16] describe the statistical outlier types as applied to nuclear material accounting. An **additive outlier** affects a single observation. It can be thought of as an abrupt removal or one time loss as a theft. In contrast, an **innovative outlier** would affect subsequent observations because of serial correlation. It indicates leakage or systematic theft over a number of periods (trickle loss). Early identification of an outlier and its type would obviously minimize loss since the organization has additional information regarding the form of material loss to confront the problem quickly, find the cause of the problem, and to solve it more effectively.

Prasad et al.[19] recently tested the **joint estimation procedure,**[37] an extension of the GM idea, with satisfactory results on nuclear material inventory data sets with known outliers and assignable causes. It can deal with deviations from any type of time series model of a process, not only AR(p). It is also more robust and discriminating in identifying outlier types, using standard hypothesis testing—not only additive (an abrupt removal or one time loss) and innovative (a continuing loss of short duration) but also **temporary change** (a continuing loss of longer duration) and **level shift** (a long term protracted removal or continuing loss throughout the remaining series).

In this section we have emphasized Nuclear Materials Accounting to demonstrate a unique application of outlier detection to a crucial problem of society. The GM procedure, the influence function matrix, the M-type iterative procedure, the joint estimation procedure, data bounding, and polynomial smoothing have also been successfully tested on conventional industrial process control data sets with known outliers.[7,20-22]

C. TWO NEW ALGORITHMS FOR THE DETECTION OF OUTLIERS

We have developed outlier detection methods which also utilize the time series pattern to model the underlying process but are simple in concept and flexible. The concept is that forms of data bounding and smoothing filter out much of the random errors, noise, and outliers in the original data. What remains should be essentially the underlying time series pattern. The smoothed or adjusted curve represents the underlying process model. In contrast to this adjusted graph, the outliers or potential loss points in the original data should be more obvious and thus easier to detect both quantitatively and qualitatively. Although especially appropriate for autocorrelated data sets, these methods can be used on any time series. With the proper fine-tuning of the sensitivity constants of these methods and consequently the sensitivity, the user can choose the method which is more suitable to the particular process conditions and environment.

We successfully tested polynomial smoothing[26-30] and data bounding[29] for process control for SPC of a chemical process.[22] Since their descriptions, computer implementations, results, and discussion are detailed in a previous issue of this journal,[32] the interested reader may refer to it. In this paper, the scope is expanded to the detection of nuclear material losses.

The first method, polynomial smoothing, is a commonly used smoother combined with our "outlier detection procedure for process control". The second method, data bounding, is essentially a means of adjusting data points that lie beyond certain limits, thus smoothing significant peaks and troughs. We developed a variation and adapted it to outlier detection.

Each method has its own independent nomenclature, although N_{drop}, Z_3, and Z_4 are common to both of them. The sensitivity constants N_{drop}, Z_1, Z_2, Z_3, Z_4, m (the number of moving points), and the degree of polynomial fit are chosen by the user. They will be referred to as sensitivity constants in contrast to parameters which are population characteristics that are generally estimated (e.g., the process mean and the standard deviation). Also common to both methods are the trimmed estimators,[23,24,39] modified process average (MPA) and the modified standard deviation (MSD) as seen in the following formula:

$$\text{MSD} = \sqrt{\sum_{i=1}^{N-N_{\text{drop}}} \frac{(X_i - MPA)^2}{N - N_{\text{drop}} - 1}} \tag{3}$$

The center line of our modified control chart is set equal to the expected process mean as planned or designed, i.e., a fixed target value (TV). In the case of a nuclear material balance (MB) or material unaccounted for (MUF), TV = 0 as opposed to MPA which is seldom zero.

Polynomial smoothing and other methods which utilize the outlier detection procedure for process control indicate the basis for the identification of each outlier in their output as follows:

1 = outlier on the original data (X_t) first detected as part of a significant peak above the adjusted or smoothed point (Y_t) and TV.

2 = outlier first detected as a significant trough below Y_t and TV.

0 = outlier found only becase of the TV ± 3(MSD) limits and does not meet the criteria of either 1 or 2.

Data Bounding for Process Control. This algorithm is based upon the deviation of each data point (X_t) from the average ($\bar{X}_{(i)}$) of its adjacent points, where $i = 1$ refers to the past two adjusted points and the next two raw data points and $i = 2$ refers to the average of the preceding three adjusted points. If the data point (X_t) is beyond $\bar{X}_{(1)} \pm Z_1$(MSD) or $\bar{X}_{(2)} \pm Z_1$(MSD) in the appropriate direction, it is operationalized as an outlier.

The output indicates the basis for the identification of each outlier as follows:

1 = outlier first identified using the average of the four adjacent points—the preceding two adjusted points and the next two raw data points.

2 = outlier first identified using the average of the three preceding adjusted points.

3 = outlier first detected according to both no. 1 and no. 2.

0 = outlier found only because of the TV ± 3(MSD) limits and does not meet the criteria of either 1 or 2.

Fine-Tuning the Sensitivity. Although reasonable default values for the sensitivity constants are already set in the algorithms, further sensitivity adjustments are often necessary, depending upon the process conditions, organizational needs, product specifications, the relative costs of a false alarm vs overlooking an outlier, the probability of a diversion, and the probability of discovering a diversion (1 − β).[11] There are several principles and guidelines which should be helpful in this empirical procedures.[22,32] After the modified standard deviation (MSD) is established, the sensitivity constants should be initialized by using, if available, a data set with known outliers, i.e., nuclear material losses or known removals by experiment. This may serve as a training set.

Bissell[23] advocates the use of a training set, which he calls a "trial control chart", to establish the target value and the control limits of conventional $\bar{X}$, M, R, and s charts. His procedure assumes that there is little or no information about the data set and the process may or may not be in control. The target value is set equal to either the process average or the midspecification value and then a trimming procedure is used in the estimation of the parameters. Consideration is given to changing the choices of the sensitivity constants to match new levels of performance and greater experience with the process. Some of the same principles are used in our parameter estimates and the fine-tuning of the sensitivity constants in the outlier detection methods.

We note that it is difficult to ascertain when the process is truly in control[5] due to, for example, fluctuations in the process mean or unknown trickle losses in the case of nuclear material balances. Thus having a training set is very helpful but not indispensable.

Since the cost of overlooking an outlier is very high in the case of nuclear materials, management may tolerate a certain frequency of false alarms. At the very least, a questionable outlier should serve as an early warning that a loss may be occurring. Management should be alert, and the operator should perform at least a cursory examination of the critical points in the process, look for a trend, and then monitor the process more closely.

D. TESTING THE ALGORITHMS ON DATA SETS WITH KNOWN DIVERSIONS

To obtain a more definitive test, we used nuclear material balance data sets with known outliers (losses), i.e., actual out-of-control nuclear inventories due to known protracted removals of nuclear material.

A series of experiments were conducted in 1980 at the Allied-General Nuclear Services (AGNS) Barnwell Nuclear Fuels Plant in order to improve the detection of nuclear material losses in the different unit process accounting areas (UPAAs). The material balance data were recorded periodically on a near real-time basis with the aid of a computerized nuclear materials control and accounting system.[34]

The experiments essentially involved physically removing nuclear material in different forms from a plutonium purification process (PPP) with variations in what was included in the UPAA for the material balance measure, location of removals, time, quantity, concentration, and type of diversion. The process includes pulse columns (2AP and 3AP), a surge tank (1BP), and strip columns (2BP and 3BP). The original researchers called these experiments "miniruns".

Table 1[19] gives a summary of the location and timing of removals for the minirun data sets. In these particular experiments, solutions of uranium separated from the plutonium are gradually removed from different tanks (i.e., protracted removals or trickle losses), and material balances are taken every hour across the corresponding UPAA.

Ideally, trickle losses should be detected in the observations immediately following the starting time and continuing through the observation immediately following the ending time according to column 5, "obs." The total protracted removal across the extended time period by volume of solution and the corresponding uranium content is recorded in columns 6 and 7.

During the protracted removal periods, it is very doubtful that a negative MUF outlier can indicate a removal or loss. On the contrary, a negative MUF outlier should indicate a release of material held up somewhere in the process as caking in the tubes or precipitated solids returning to solution. Of course, measurement errors are common. If the preceding inventory measure (I_{t-1}) is erroneously low (see eq 2), MUF_{t-1} will be too high. However, if I_t is accurate, the next observation (MUF_t) will be too low. We now consider particular miniruns.

Table 1. Location and Timing of the Removals for Four of the Minirun Experiments

experiment & location	tank	start time/date	end time/date	obs.	total removed vol. (L)	total removed U (Kg)
Minirun 3C		0815/07/18	1204/07/21			
	2AP	1645/07/18	0845/07/19	8−24	96	4.224
UPAA PPP	3AP	1645/07/18	1645/07/19	8−32	96	4.253
	1BP	1615/07/20	0850/07/21	56−72	96	5.722
U settled on bottom of acid concentrators = 120 Kg						
Minirun 4A		0700/09/04	1900/09/07			
UPAA 1BP	1BP	0615/09/06	0545/09/07	48−71	299.4	17.64
Minirun 4B		0700/09/04	1200/09/08			
	3BP	1141/09/04	0445/09/06	4−45	370.3	20.99
UPAA column	1BP	0615/09/06	0545/09/07	47−70	299.4	17.64
& sample Tk	2BP	0900/09/07	1045/09/08	74−99	826	22.1
U settled on bottom of acid concentrators = 100 Kg						
Minirun 5B		1600/11/18	1630/11/21			
	1BP	0000/11/19	1200/11/19	9−20	174	10.2
UPAA column	1BP	1445/11/19	0115/11/20	23−34	177.2	11.1

Minirun 3C. Tables 1 and 2 show that there are three protracted removals from three tanks in this experiment, two of which are in part simultaneous: 0.4186 Kg/hr during periods 8−24; 0.1701 Kg/hr during periods 25−32; and 0.3366 Kg/hr during periods 56−72. The 120 Kg that settled out in the concentrators averages to 1.58 Kg/h. Thus the modified process average (MPA = +1.145 Kg) and median = 1.65 Kg are above the target value (TV = 0), but they should be even higher because of those known removals and precipitation. Over the entire period, the separated solids (120 Kg) and the amount removed (14.199 Kg) or 134.2 Kg total should give an average MUF = 1.77 Kg/h over the 76 periods.

Data bounding (Figure 1) and polynomial smoothing detected a similar number of losses. The summary in column 3 at the bottom of Table 2 shows that

TOI = total outliers identified = 13

T+OI = total positive outliers identified = 7

NA+I = number of actual positive outliers identified = 4

NFA = number of false alarms = 3

N+OM = number of positive outliers missed = 22

Both methods gave T+OI = 7. Of these, the number of actual positive outliers identified NA+I = 4, which is 15.8% of the positive observations during the protracted removal periods. The difference,

$$\mathrm{T{+}OI} - \mathrm{NA{+}I} = \mathrm{NFA}$$

or

$$7 - 4 = 3$$

the number of false alarms, which is 14.3% of the positive observations during the nonremoval periods. The number of known positive outliers during the protracted removal periods (26) less the number of actual positive outliers identified (NA+I = 4) gives the number of positive outliers missed (N+OM = 22).

It is crucial to detect an outlier as early as possible within the protracted period of removals. Data bounding and polynomial smoothing detected the very first observation (no.

Table 2. Minirun 3C—UPAA PPP (2AP, 3AP, 1BP)

Detection of Outliers among Uranium Inventory Differences (MUF)—AGNS Barnwell Nuclear Plant
* = detection of outlier
*+ = detection of outlier beyond TV ± 3(MSD), using data bounding or polynomial smoothing
$N = 76$
min = −40 Kg, max = 15.1 Kg — target value (TV) = 0
modified process average (MPA) = 1.145 — median = 1.65
modified standard deviation (MSD) = ±3.571 — % drop = 5% (4)
total no. of observations during removal periods = 42
total no. observations during nonremoval periods = 34
no. of + observations During removal periods = 26(61.9%)
no. of + observations during nonremoval periods = 21(61.8%)
amount of uranium that settled out = 120 Kg = 1.6 Kg/h

time	MUF	DB-III	poly smooth	joint estim	CUSUM	assigned cause
4	−6.5	*3	*2			none or unknown
8	5.5	*3	*1			protracted
10	−8.0	*3	*2	*3		0.4186 Kg/h
13	1.8					removal
14	4.7			*1		obs. 8−24
15	4.4					17 periods
16	6.2					" "
17	2.8					" "
18	5.2					" "
19	6.7	*1	*1			" "
20	3.1					" "
22	1.1					" "
23	3.1					" "
24	1.3					" "
25	5.3	*1	*1			0.1701 Kg/h
26	0.3					obs. 25−32
29	−7.8	*3	*2	*2		8 periods
30	0.3					" "
31	1.9					" "
37	5.4	*3	*1			none or unknown
39	7.2	*3	*1	*1		" "
42	6.6	*3	*1			" "
45	7.0			*1		" "
56	0.45					protracted
58	−5.0	*3				removal
59	3.0					0.3366 Kg/h
60	1.8					obs. 56−72
61	15.1	*+3	*+1	*2	*	17 periods
62	−5.2	*3	*2			" "
63	3.6				*	" "
64	2.4				*	" "
65	3.6				*	" "
66	−1.0				*	" "
67	1.4				*	" "
68	1.6				*	" "
69	−2.3				*	" "
70	0				*	" "
71	0.7				*	" "
72	−40.0	*+3	*+2	*2	*	" "
74	−5.2			*1		none or unknown
total	(TOI)	13	12	8	11	
total	(T+OI)	7	7	4	7	
actual	(NA+I)	4(15.4%)	4(15.4%)	2(7.7%)	7(26.9%)	
false	(N+FA)	3(14.3%)	3(14.3%)	2(9.5%)	0	
missed	(N+OM)	22	22	24	19	

data bounding III: $Z_1 = 1.1$; $Z_3 = 3$; $Z_4 = 1.4$
polynomial smoothing (Quad-7): $Z_2 = 0.7$; $Z_3 = 3$; $Z_4 = 1.4$
In column 5—i.e., the joint estimation procedure:
1 = temporary change, 2 = additive outlier
3 = innovative outlier, 4 = level shift

8) in the initial protracted removal period. The joint estimation procedure missed that one, but did detect no. 10 which is negative and the positive no. 14. It also had a greater incidence of false alarms.[19] In the final protracted period (nos. 56−72), data bounding detected the negative no. 58, and all three methods identified the positive no. 61. CUSUM missed the initial protracted removal period completely but did very well on the final removal period.[34]

Ideally, any comparison of results among different methods should be based upon equal sensitivities. To date, the methods proposed herein do not have easily quantifiable sensitivities since at least three sensitivity constants are used

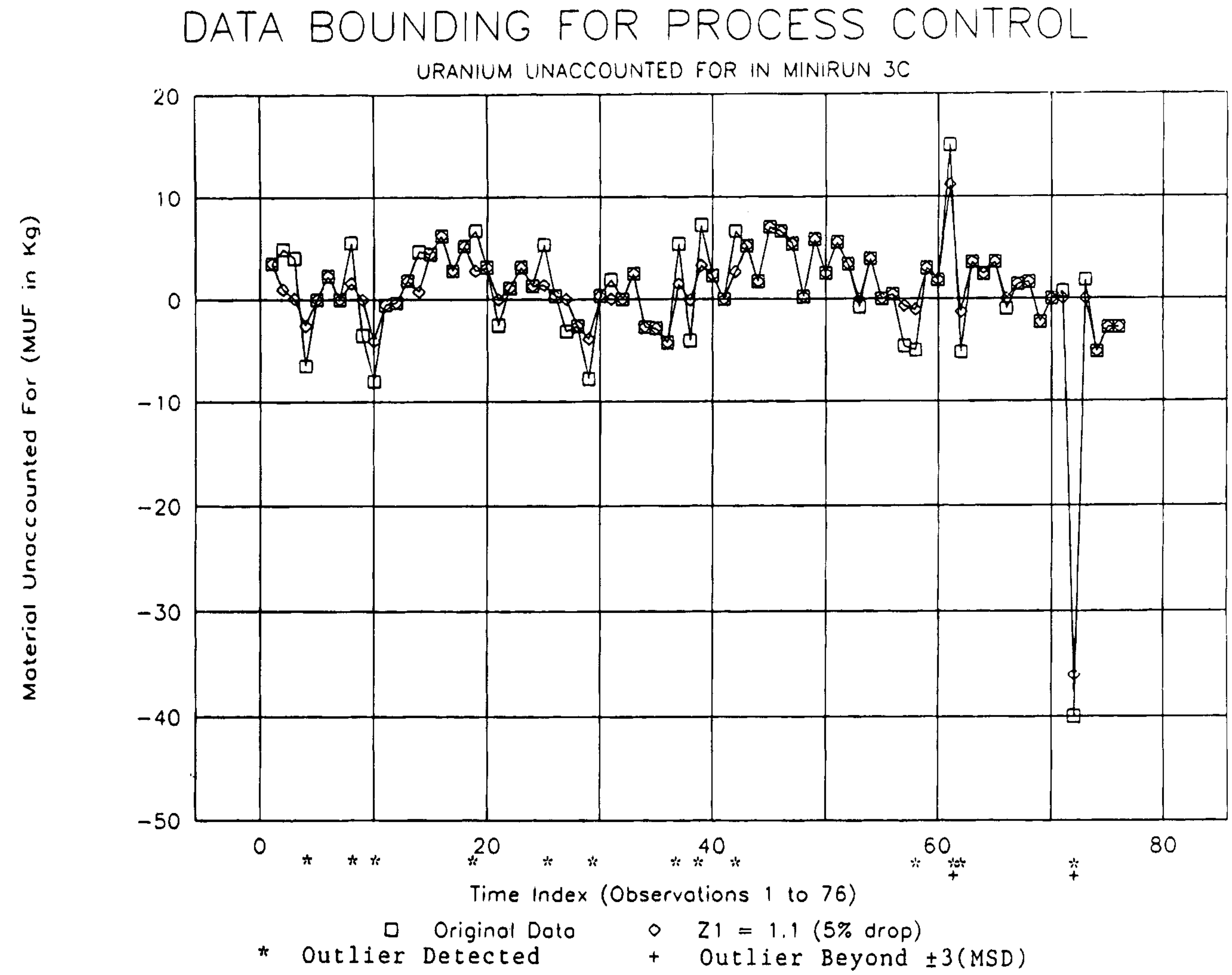

Figure 1.

in each method. Thus the different methods are not exactly comparable, although the comparisons do give some idea of the relative performances.

Minirun 5B. This run has two protracted removal periods that are amost consecutive with only a 2-h interruption separating them. Both are from the surge tank (1BP): 10.2 Kg (0.850 Kg/h) during observation nos. 9–20 and 11.1 Kg (0.925 Kg/h) during period nos. 23–34. The nonremoval period has a much greater proportion of positive MUFs (59.1%) than the protracted removal periods (37.5%), making it very difficult to detect true losses while minimizing false alarms.

Although data bounding and polynomial smoothing (Figure 2) immediately detected a negative outlier at point no. 9 in the first protracted removal period (Table 3), they did not detect a positive outlier until no. 17 and no. 18. The joint estimation procedure only detected negative outliers at observations no. 19 and 20;[19] but again, the sensitivities may not be comparable to the proposed methods. All three methods did not detect any loss in the second removal period (nos. 23–34) until near the end at point no. 33. Again CUSUM was not useful since it detected 56 consecutive outliers from observation nos. 3–58, well before and past the removal periods.[34] The results of minirums 4A and 4B together with the four complete minirun data sets may be found in Sebastian.[22]

Analysis. The outliers in these data sets are very difficult to identify because each removal per period is so small and generally does not exceed the normal in-control chance fluctuations. In fact, the removals are usually within 0.2-(MSD) of the target value.

The percentages of positive MUF observations during the removal and nonremoval periods are quite similar, making it very difficult to detect losses. In minirun 3C (Table 2), for example, there is often little difference between the removal and nonremoval periods, i.e., 61.9% of the observations were positive during the removal periods vs 61.8% during the nonremoval periods. In miniruns 5B, there is a greater frequency of positive MUFs during the nonremoval periods.

Tables 2 and 3 show that the percentage of positive outliers detected by data bounding and polynomial smoothing during the removal periods is consistently higher than during the nonremoval periods. The same was found for miniruns 4A and 4B.[22] Only in minirun 3C is it close—15.4% vs 14.3%. The joint estimation procedure did not do as well, but that may be because of different sensitivity settings.

It must be remembered, that although the three methods adapt to the process change as if it were a new process, the joint estimation procedure does this to a greater degree. It often will detect only the first outlier during a protracted removal period and when the process reverts to its normal

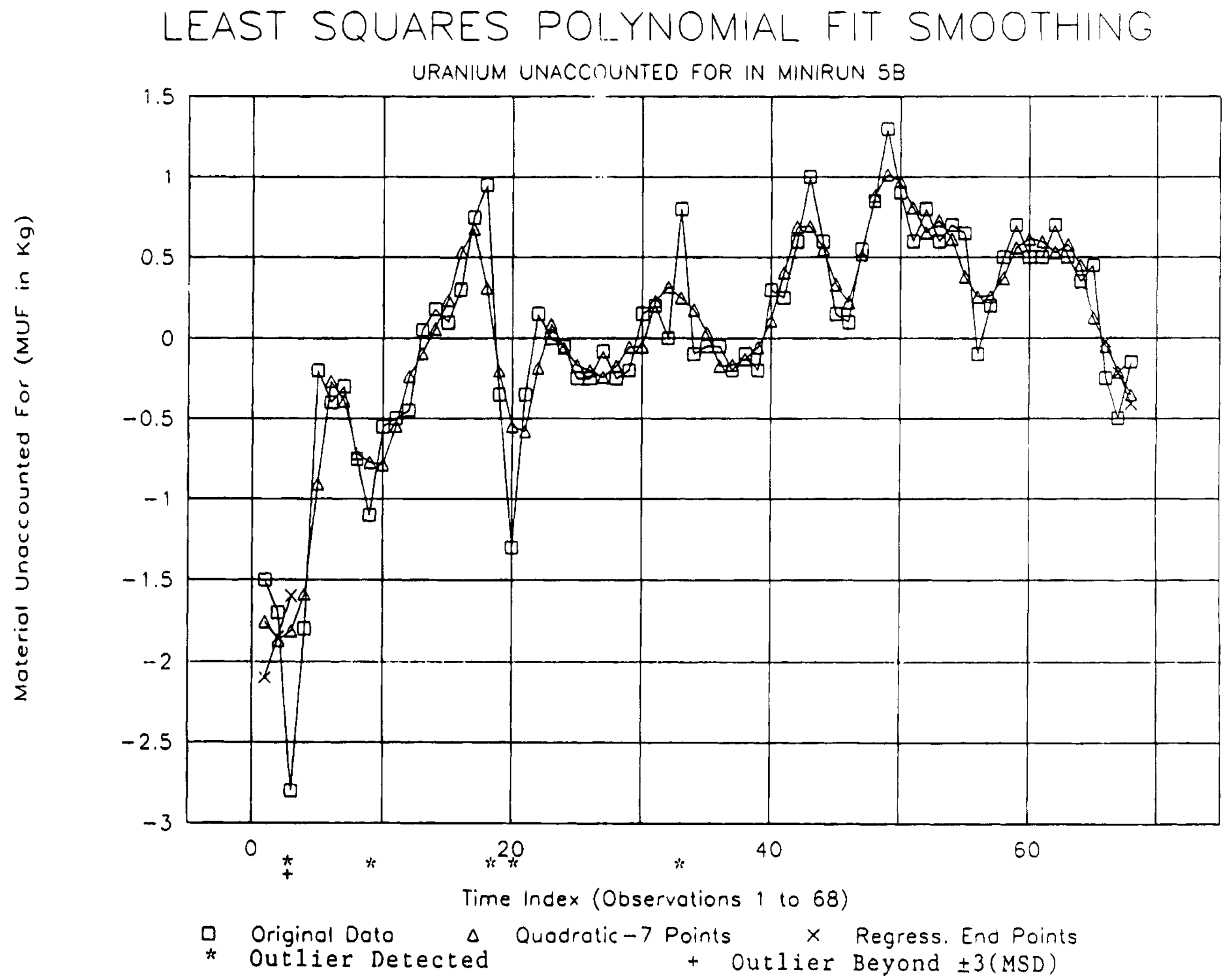

Figure 2.

state, it may also detect that change as an outlier. The joint estimation procedure did, however, have a lower incidence of false alarms.

CUSUM. Each material balance is taken over each interval, i.e., two successive observations. Thus previous cumulative losses are not considered, and a small protracted removal per hour may go unnoticed because they are combined with large normal fluctuations. Therefore the CUSUM method may be used as a check. However, minirun 3C shows that it may take many periods of a protracted removal before CUSUM can detect any loss. In miniruns 4B and 5B, there were so many consecutive outliers detected as to be almost meaningless. A material balance over a large number of periods may also serve as a rough check.

Precipitated Solids. In miniruns 3C and 4B, outlier detection is further complicated by the fact that 120 and 100 Kg, respectively, of uranium solids settled to the bottom of the concentrators. If there is a steady state or equilibrium, that would be no problem, but new material is constantly entering the process. Since these solids apparently were not considered in the nuclear inventories, positive MUFs or losses would be recorded.

We must assume that the original researchers conducted the experiments properly and removed the solids from the concentrators before the beginning of each run. But there is no way of knowing at what rate the particles settled. If constant, the uranium precipitated out of solution at a rate of 1.58 Kg/h or period in minirum 3C and 1.0 Kg/h in minirum 4B. This greatly exceeds the rate of protracted removals which range from 0.170 Kg/h to 0.735 Kg/h.

Thus there may be some confusion as to whether an identified outlier represents a bona fide loss (removal) or simply solids that precipitated out. Therefore, a number of false alarms might be expected. At the same time, however, the methods should be able to adapt to the settled out solids if the rate of precipitation is relatively constant as part of the process.

Table 4 compiles the average solids settling per hour and the average amount of material removed per hour over the duration of each experiment. The sum of these gives an expected average MUF.

It can be seen that the modified process average (MPA) is consistently less than the expected average MUF, leading us to suspect a systematic measurement error. Only in minirun 4A is there a known measurement problem, i.e., the plugged probes.[22]

Summary. All of the methods had difficulties in detecting the known outliers because of the very small removals and other problems that were discussed. In comparison to the other methods, data bounding and polynomial smoothing did quite well, but no definitive conclusion as to relative performance can be made because of the different sensitivities that each method used. In addition, the joint estimation procedure is more geared to detecting the first outlier of an

Table 3. Minirun 5B—UPAA Column (1BP)

Detection of Outliers among Uranium Inventory Differences (MUF)—AGNS Barnwell Nuclear Plant
* = detection of outlier
*+ = detection of outlier beyond TV ± 3(MSD), using data bounding or polynomial smoothing
$N = 68$
min = −2.80 Kg, max = 1.30 Kg — target value (TV) = 0
modified process average (MPA) = 0.040 — Median = 0.075
modified standard deviation (MSD) = ±.613 — %drop = 2%(2)
total no. of observations during removal period = 24
total no. observations during nonremoval period = 44
no. of + observations during removal periods = 9(37.5%)
no. of + observations during nonremoval periods = 26(59.1%)

time	MUF	DB	poly smooth	joint estim	CUSUM	assigned cause
3	−2.8	*+3	*+2		*	none or unknown
4	−1.8	*1			*	" "
9	−1.1	*3	*2		*	protracted
13	0.05				*	removal period
14	0.18				*	0.850 Kg/h
15	0.1				*	obs. 9−20
16	0.3				*	12 periods
17	0.75	*2			*	" "
18	0.95	*3	*1		*	" "
19	−0.35			*3	*	" "
20	−1.3	*3	*2	*2	*	" "
30	0.15				*	removal period
31	0.2				*	0.925 Kg/hr
33	0.8	*3	*1	*2	*	obs. 23−34
43	1.0	*3			*	none or unknown
48	0.85	*2			*	" "
49	1.3	*3			*	" "
58	0.5					" "
total	(TOI)	10	5	3	56	
total	(T+OI)	6	2	1	28	
actual	(NA+I)	3(33.3%)	2(22.2%)	1(11%)	9(100%)	
false	(N+FA)	3(11.5%)	0	0	19(73.1%)	
missed	(N+OM)	6	7	8	0	

data bounding III: $Z_1 = 0.9$; $Z_3 = 3$; $Z_4 = 1$
polynomial smoothing (Quad-7): $Z_2 = 0.5$; $Z_3 = 3$; $Z_4 = 1$
In column 5—i.e., the joint estimation procedure:
1 = temporary change, 2 = additive outlier
3 = innovative outlier, 4 = level shift

Table 4. Expected Average MUF vs MPA (Kg)

experiment	settled solids	av settled	total removed	av removed	expected av MUF	MPA
minirun 3C	120	1.58/h	14.199	0.187/h	1.77	1.145
minirun 4A			17.64	0.210	0.210	−0.278
minirun 4B	100	1.00	60.73	0.607	1.61	1.08
minirun 5B			21.3	0.313	0.313	0.040

adverse process change as it adapts to what is essentially a new process.

E. DISCUSSION

Other more outlier resistant smoothing methods—e.g., EDA smoothers[35] or a LOWESS smooth[36]—may improve outlier detection. Such nonparametric smoothers give larger residuals between the outliers and the smoothed data which represents the underlying process model. These smoothers are currently being tested in our laboratory.

If there are protracted removals or if there are consecutive outliers—i.e., an uncorrected adverse process change over several observations—data bounding and polynomial smoothing tend to model the process change as is. Thus they may not detect some of these outliers relative to the original in-control time series pattern. They will certainly be detected if beyond TV ± 3(MSD).

Polynomial smoothing and to some extent data bounding adjust to an out-of-control situation as though it were a new process especially in the case of consecutive outliers. Then the primary operationalization condition may only detect the beginning of a protracted removal or leakage situation, i.e., the first outlier as the joint estimation procedure does.[37,19] When the leakage is stopped and the process is brought back into control, there is another process change back to the original conditions and the next point may appear as an outlier. Since the other outliers within the adverse process change period may be undetected, the secondary operationalization condition is indispensable to keep β low.

Neither method developed herein can identify the type of outlier directly. The GM, M-type iterative, and the joint estimation procedures utilize complex statistical mathematics to do that after mathematically modeling the process.

Real-Time On-Line Process Control Capability. The results may give the impression of applying only to retrospective control. Because the algorithms are designed to handle end points, they also apply to real time on-line process control, i.e., as soon as the latest measurement is made.

In data bounding, the reader may note in Tables 2 and 3 that any outlier identified with a +*, *2, or a *3 notation would have been detected if it were a final point. All identified outliers with assigned causes except two met at least one of these criteria.

If an outlier is found by both the primary and secondary operationalization conditions as indicated in the computer output, the practitioner can feel more assured that X_t is indeed an outlier and not a false alarm. Data bounding has two primary operationalization conditions that may reinforce each other. Using both end point manipulation procedures in polynomial smoothing can serve a similar function.

Another method may confirm the indications of one of our methods or vice versa to minimize α when a false alarm is very costly. Similarly, a second method whether ours or another could be used as a means of outlier detection to minimize β if the cost of overlooking an outlier is crucial. Thus the operations manager would have a greater body of evidence for concluding an adverse process change in the way of a material loss.

Summary. We have seen that the methods used in this paper are effective across different applications but simple in concept, flexible, and adaptable to the process environment. Thus they may be more acceptable to practitioners than the more sophisticated methods mentioned in section B. The approach is clearly useful in a variety of applications, particularly in the case of autocorrelated data and most likely with smoothers such as LOWESS to severely non-normal distribution situations, often due to the presence of many outliers. The exact conditions are being determined in current research. The proposed methods adequately handle the end points, thus being able to detect outliers as soon as the latest data become available.

The most important feature of these methods, however, is the ability to identify outliers, i.e., adverse process changes, earlier than traditional methods (e.g., control chart), well within the usual three standard deviations from the target value. Thus we propose that these methods are a useful addition to those already available to operating managers involved in nuclear material safeguards.

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