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A Practical Guide to Measurement Control Experience on Non-destructive Assay Equipment at the Los Alamos National Laboratory Plutonium Facility

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ABSTRACT

If you are looking for the elegant application of statistical approaches or a sophisticated analysis of a measurement control system, read no further. However, if you are interested in experiences developing, implementing, modifying, and living with a practical measurement control program for non-destructive assay equipment, please continue. This paper goes through the basis for measurement control at the Los Alamos National Laboratory (LANL) Plutonium Facility, including the philosophy, types of safeguards, accountability instruments used for quantitative assays, frequency of tests, statistics used, the link to the LANL accountability system, management structure, how measurement control works daily and monthly, common causes of failure, deficiencies, and future upgrades.

I. INTRODUCTION

The Los Alamos National Laboratory (LANL) Plutonium Facility, located at Technical Area 55 in Los Alamos, New Mexico, is funded by the Department of Energy's (DOE) Defense Programs Division. Because this facility works with nuclear materials, it is subject to the DOE orders covering safeguards and security.

The DOE order on safeguards[1] specifies compliance-based requirements that address various aspects of measurement and measurement control for all instruments and techniques used to both qualitatively and quantitatively measure nuclear materials under safeguards in the Plutonium Facility and all DOE facilities.

The LANL Plutonium Facility works with a wide variety of nuclear material in gaseous, liquid, and solid forms. Depending on the type and quantity of nuclear material present, these materials are valuable and hazardous. The value relates to both the cost associated with producing these materials, as well as their ability to be used in nuclear weapons. To properly deal with the associated value and hazards, including toxicity, radiation, and criticality, knowledge of the quantity and isotopes in each item is of paramount importance.

The least hazardous and most cost-effective method to quantify amount and type of nuclear material in a container is through the use of non-destructive assay (NDA) equipment. This equipment is designed to base a quantitative measurement on the radiation emitted by the isotopes associated with nuclear material. This is the primary quantitative approach used by the Plutonium Facility.

Measurement control is so fundamental that even without any DOE order or other regulations requiring measurement control, it would be employed by anyone performing defensible measurements. This does not mean that the measurement control system would be identical to one under compliance-based standards, but that the principles will be the same.

At LANL we define a defensible measurement as one that can be demonstrated to be from instrumentation or techniques operating within their desired levels of bias and precision.[2] Here we define bias as the difference between the average measured value and the true or reference value. Percent relative bias is defined as the bias expressed as a percent of the average value. Precision is defined as either the standard deviation of measurements on the same item, or the percent relative standard deviation depending on the context.[3]

It should be noted that compliance-based requirements, are not necessarily either technically correct or the most efficient approach. It seems to be an unfortunate reality of regulations that once issued, regulations live forever regardless of inaccuracies, unnecessary rigidity, or the best of intentions to correct them during the next revision. Clearly there is no intention to be incorrect. Nevertheless, we have seen requirements that are prescriptive to the point of not being

applicable, and once this occurs, there may be situations that demand, from a technical or efficiency standpoint, non-compliance. Obviously, this is a very uncomfortable position. We have had such situations and have successfully defended our position simply because we had a technical basis to do so. However, this also demands knowledgeable external and internal auditors who can understand the situation.

II. WHY IS MEASUREMENT CONTROL DONE?

The primary function of any measurement control system is to assure that an instrument is functioning within its desired level of bias and precision whenever it is used. However, other benefits can be gained from this system as well. Examples of other benefits include understanding the instrument's capability in a different or changing operational environment, improving the instrument's capability, understanding the process that generates the items for measurement, and knowing/defending your inventory. These are all of critical importance to the Plutonium Facility. Some functions, depending on facility-specific applications, may well be more important than the primary function.

For example, the LANL Plutonium Facility became operational in 1978. This predates the current extensive use of NDA equipment. Because NDA was not included in any facility design consideration, most instruments are located in areas with less than ideal measurement conditions. Inadequate shielding, combined with the storage and movement of significant quantities of special nuclear material (SNM) during staging to and from NDA instruments, results in a fluctuating high radiation background. Consideration of this background not only influences the impact on the instrument's bias and precision (which in combination will be referred to as "uncertainty"), but influences other considerations such as the most appropriate NDA technique given the operational environment. The need for improved background correction or shielding techniques can be revealed through measurement control data analysis.

As a research and development facility for plutonium, the processing systems are, by design, constantly under change. To understand such processes and maintain knowledge on mature processes requires monitoring nuclear material throughput. This in turn requires defensible measurements to draw a mathematical balance around a process to detect unusual changes in throughput.[a] It is the measurement control program (MCP) in association with the calibration program that assures that the measurements and their uncertainty are well known. This is particularly important for throughput and propagation of variance calculations.

To emphasize this point, the MCP must also address the uncertainties associated with all measured items. This is a major dilemma, especially at a research and development (R&D) facility. The ideal state for any NDA instrument would be to have standards that matched all matrices to be measured on the instrument. It would then be a simple matter to determine the bias and precision for each matrix. However, reality could not be further from this ideal condition. In our R&D facility we not only have multiple matrices (currently over 80 different types), but these are often composed of a small number of items and new items can be generated with high frequency. The cost associated with trying to make standards for each matrix would be prohibitive. In addition, the variation in some matrices, especially waste, can be so variable that each one is unique.

Faced with the daunting problem of trying to assess the precision and bias for each matrix, we have tried to address this problem through the use of what we term a "remeasurement database." To generate data for this database, we deliberately remeasure items as part of our inventory control program. This database is composed of two parts, precision data and bias data. The first compiles repeat measurements on the same items grouped by similar matrices by the same technique. The second compares the results for the same items by different techniques. We are fortunate at the Plutonium Facility to have a large suite of instruments. Part of that suite includes calorimeters. With the large measurement chambers available in our calorimeters and the high accuracy of this technique, calorimetry provides a critical evaluation of bias on other less accurate techniques.

Lastly, to maintain an effective criticality program and to defend the facility's inventory during audits, a measurement control program provides the foundation to demonstrate that the NDA measurements are defensible.

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[a] This balance is commonly referred to domestically as the inventory difference (ID) or internationally by the International Atomic Energy Agency (IAEA) as material unaccounted for (MUF), and can be either positive or negative.

III. PHILOSOPHY

Regardless of the initial philosophy used to design a system, over time and with experience it is not uncommon for this philosophy to evolve. It is important to allow flexibility in any system to address internal improvements, as well as those driven by external forces such as, in our case, new DOE requirements.

The current measurement control program was initiated in the early 1980s. The fundamental philosophy has changed little since then and includes the following main elements.

1. A computer-based measurement control system is used to perform short time frame tests on NDA equipment and maintain a database for long time frame trend analysis.
2. The MCP is tied into the safeguards accountability system to prevent data entry from instruments not currently passing the MCP.
3. For each generic instrument/technique a measurement control failure response plan is developed and implemented to assure consistency and completeness in response to a MCP failure.
4. After a measurement control failure, at a minimum, all items measured since last passing measurement control are suspect.
5. A short time frame measurement control test is used with failures based on 2-sigma and 3-sigma limits.
6. The instrument operator can resolve 2-sigma limit failures directly by remeasurement.
7. Three sigma limit failures can only be resolved by the operator's input to an independent group indicating that the instrument is in statistical control.
8. A long time frame test is used that performs trend analysis and checks against desired levels of bias and precision.
9. A record tracking system responds on an instrument-specific basis and documents all actions for lessons learned and for future review during audits.
10. Where practicable, the system meets the intent of regulatory requirements.

One factor, which directly led to a computer-based system, was the large number of instruments to be addressed by this system. This will be discussed in section V.

At our facility, element number four results in the remeasurement of all items since the last measurement control failure. Because our measurement control tests are conducted at a short interval, this is not a great burden on our systems. If, however, a longer interval is chosen, or due to short measurement times the number of items is large, other remeasurement strategies can be considered under the same philosophy.

IV. HOW TO START: BASIS FOR LIMITS

Ideally, the first step is to determine the desired precision and bias required for each isotope/matrix to be measured. This is followed by selection of an instrument(s) that meets the desired performance. Besides the obvious requirements of meeting defined measurement uncertainty limits for desired isotopes, the influence of the operating environment must also be considered to assure all requirements can be met. However, often an instrument exists and a new matrix is either generated or received by the facility. It is then the operator's responsibility to assess if the existing instrument has the ability to measure the new matrix. At LANL each instrument must go through a certification protocol. This takes place after calibration and satisfactory operation and before use as an accountability instrument.[4] This protocol has the following elements.

1. Use a statistically based data collection plan on standards as similar to the process items to be measured as possible (see Table 1, Instrument Stability Measurement Matrix).
2. Collect data at place of use and under actual operating conditions.
3. Measure National Institute of Standards and Technology (NIST) traceable or well-known standards that span the range of instrument calibration.
4. If more than onee measurement technique is available for the same items, collect comparative measurements of actual process items.

The statistically based data collection plan in element one assures randomization of the sequence in which standards have been measured. This approach allows for meaningful estimates of the standard deviation.

Table 1. Instrument Stability Measurement Matrix

	M	T	W	T	F
AM	BKG*	BKG	BKG	BKG	BKG
AM	STD1	STD6	STD1	STD3	STD3
AM	STD6	STD3	STD3	STD1	STD6
AM	STD1	STD1	STD6	STD1	STD3
PM	STD3	STD6	STD1	STD6	STD1
PM	STD3	STD6	STD3	STD1	STD6
PM	STD6	STD3	STD1	STD3	STD1
PM	STD1	STD6	STD6	STD1	STD3
ASSAYS	8	8	8	8	8

*BKG = Background measurement

The potential impact of the lack of such a plan was revealed during the recent certification of a mass spectrometer. The operators wanted to use the data set they had already collected during instrument setup and testing. The request to use existing data that is not statistically based is a constant problem. Review of this data set revealed non-random daily tests in which the concentration of the gas standards being measured always increased. These operators were convinced that their instrument was ready for operation because they demonstrated uncertainties far less than the desired levels of precision and bias.

The operators were now required to repeat two weeks of measurements using a randomized design. These new tests revealed variations on the order of hundreds of percent on the low concentration gas standard whenever it was preceded by a high concentration gas standard. An improper procedure for clearing the gas measurement chamber caused this problem. Their original data collection scheme hid this deficiency.

Element two cannot be stressed enough and should be a warning to all operators who accept manufacturer-stated performance levels on their instruments without testing the instrument at place of use and under actual operating conditions. The manufacturer's specifications are probably obtained under more stable operating conditions than are found in practice.

With respect to element three, the quality of the standard depends on the use of the instrument. An instrument designed to measure high-purity metal or oxide requires higher quality standards than the same instrument if it is only used to measure waste. In reality, the difficulty in developing standards representative of waste will often lead to procuring higher quality standards due to their easier design and availability. It should also be noted that the uncertainties of standards have a greater impact as the uncertainties between standards and techniques converge, and this impact must be considered in the overall uncertainty calculations.

It is critical to have a basis to compare measurement techniques. This is the rationale behind element four. If the same item is measured by different techniques, we expect that there will be biases between the different techniques. At LANL we have many items that are in long-term storage in our vaults. They often have a measurement basis that could be from a measurement made a decade ago on an instrument that has been decommissioned and is no longer available. By incorporating items representative of the matrices previously measured into the certification of a new technique, these biases can be quantified. Unquantified biases on safeguarded nuclear material can be incorrectly interpreted as potential diversion of this material.

Assuming that the instrument has behaved in a normal manner during the certification procedure, limits for the measurement control program are derived from the data collection plan. These limits are then implemented through entry into the MCP. The instrument is now placed into service (meaning that the measurement results from this instrument can be entered into the accountability database). Because of the lack of a strong historical record on the MCP, this instrument is provisionally certified with a formal review of these limits scheduled for three months after being placed in service.

Operator training is another important aspect of operating any instrument. In the case of LANL, the subject matter expert who originally wrote the operating procedures and took the instrument through the certification process is responsible for training the technicians who will operate this instrument.

V. NON-DESTRUCTIVE ASSAY INSTRUMENTS

In section III, we mentioned that the first key element was a computer-based system for short-term measurement control tests. The decision to use a computer-based system was dictated by the large number of instruments requiring measurement control. In the case of the Plutonium Facility alone, the instruments total over 140. Some are in our NDA laboratory, noted as off-line, and some in our gloveboxes, noted as in-line. Although our measurement capability is in a constant state of flux as processes are modified, the average number of instruments per instrument type is listed below with a brief description of the operating principle for each.

1. Electronic balances—100 (majority are in-line)
   Magnetic transducer that relates movement of metal core in magnetic field to mass.
2. Segmented gamma scanners (SGSs)—3 (all off-line)
   Gamma ray-based quantitative measurement that relates gamma ray peak area for a specific radioactive isotope to mass. Multiple segments are measured per item to minimize matrix effects and the measurement results from each segment are then summed.
3. Passive and active neutron—13 thermal neutron counters (TNCs) or high-level neutron coincidence counters (HLNCCs). Seven are in-line and the rest are passive/active neutron (PAN) coincidence counters that are off-line.

Both techniques use coincidence counting of neutron events. This time correlation technique between neutron events is done to distinguish between the multiple events associated with spontaneous fission (passive) or induced fission (active) and single events not due to fission (such as alpha-n reactions). The number of coincidence events are then related to the mass of the isotopes present, either due to their contribution to the coincidences or isotopically related.

4. Calorimeters (CALs)—16 (all off-line)
   Heat output measurement which, in combination with gamma-ray isotopics, results in a quantitative measurement of mass.
5. Gamma isotopics (ISOs)—3 (all off-line)
   Gamma ray-based qualitative measurement of the isotopic percentage of the isotopes associated with nuclear material. In combination with calorimetry, it results in a quantitative measurement of mass.
6. Solution assay instruments (SAIs)—4 (all in-line)
   Gamma ray-based quantitative measurement that relates gamma ray peak area for a specific radioactive isotope to mass. This technique, used on aliquots of solution, measures grams per liter that is then applied to a larger volume of solution from which the sample was drawn.

VI. STATISTICAL TESTS FOR MEASUREMENT CONTROL

If an instrument is performing properly when a standard is repeatedly measured over time for measurement control purposes, the resulting measurements should vary randomly about the standard's reference value. It is important that each measurement control check be subject to a test for non-random behavior and that accumulated data be tested for the same. The immediate test might be simply a test to see if the measurement is close to the reference value; the tests on accumulated data might be tests of trends and biases.

There are many statistical tests that can be useful here.[5] However, not all of these tests are practical or needed for every application. You need to determine which measurement control tests will work best for your operational environment. We have chosen one real-time (immediate), computer-based test using two different criteria for use in our MCP.

Warning limit = ±2 sigma (~95.5%)
Action limit = ±3 sigma (~99.7%)

It should be noted that these criteria are also a DOE regulatory requirement for LANL. A failure response plan addresses the actions taken when one of these criteria is exceeded.

A caution on statistical tests is worthwhile here. Associated with each test is a chance of indicating that something is amiss when an instrument is performing normally. This is called a false positive. These false positive failure results must be dealt with in an efficient manner to minimize disruption in the working environment while assuring they are appropriately considered to avoid missing a true positive. Because each test has an expected false positive rate, performing more tests is not necessarily "better." Employing too many tests can seriously degrade instrument availability due to a high rate of false positives.

Our responses to exceeding the warning or action limits have evolved over time. This evolution came about through addressing the true impact of measurement control events. It concerns the distinction between events of statistical significance versus those of practical significance. For example, many of our electronic balances operate with a 1-sigma level of 0.1 g. Using the warning and action levels above would indicate an event of statistical significance whenever the measurement difference of a standard weight exceeded 0.2 g or 0.3 g (with the appropriate action to be taken for each level). However, the operator pointed out that the nuclear material in question is required to be measured to within the nearest 0.5 g.

This observation broadened to cover another common phenomenon seen in our NDA instruments. Our balances, SGS, and TNC instruments have often shown sudden small bias shifts for which there is not an obvious assignable cause. Our calorimeters show a small seasonal bias drift that appears to be related to humidity. In each case no reasonable corrective action is feasible. One could argue that humidity control is one possible corrective action for calorimetry. However, as stated previously, our facility was not designed for NDA equipment. Humidity control was never considered and would be prohibitively expensive at this time. Another possible action could be an intelligent measurement control system that would adjust for these biases by shifting the "zero" point for the 2-sigma and 3-sigma tests to operate around the bias. Again, we do not have that capability at this time. In any case even if we did, the first action is to evaluate the significance of these bias shifts.

Evaluation of these bias shifts determined that they are not of practical significance. To address this lack of practical significance and minimize the false positive failure rate, we decided to add a new feature, the "administrative limit," to our measurement control algorithms. This limit takes into consideration the practical significance of known events and is used to adjust the response of the measurement control system. It is set at the acceptable bias for each instrument, and acts as an additive factor to the calculated limits at the warning and action limits. The overall result is a MCP that addresses real-life operating conditions, minimizing false positive events while assuring that the instruments remain in control.

In addition to the short-term computer-based test, we also employ long-term testing. LANL's Statistics Group performs a monthly measurement control analysis for discussion at a quarterly meeting, or sooner should the data warrant it. The primary data for this meeting is based on MCP plots generated by the Statistics Group. Besides being experts in their field, they lend a level of independence to balance operational decisions. Samples of trending plots and a discussion of their contents follow.

VII. MEASUREMENT CONTROL CHARTS[b]

Accuracy test data sorted by instrument is sent monthly to the Statistics Group for analysis and plotting. The result is a statistical process control (SPC) chart for each instrument and the Instrument Performance Report (IPR). The control charts are basically X-bar - S charts that are in wide use in industry. Here however, our process is a measurement process rather than a physical product.

Each chart consists of two plots describing the performance of an instrument over the past year. The upper plot represents the closeness of the average accuracy test result to the reference value and is referred to as the "accuracy plot." The lower plot reflects the instrument variation and is called the "precision plot."

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[b] The control chart description will be given in terms of NDA instruments, excluding balances. For the NDA instruments it is assumed that the measurement error is proportional to the value of the standard. Thus, we consider the relative difference between the measured value and the standard value rather than the absolute difference. For electronic balances it is assumed that measurement error is basically independent of the value of the standard. Thus, for balances the difference between the measured value and the standard value, not the relative difference, is considered.

The measurement control accuracy test data results, expressed as the percent relative difference (%RDIF) between the measured value and the standard value, are collected and grouped by weeks or months for each instrument[c] (For electronic balances, the gram difference [DIF] is considered rather than the %RDIF). Values that are judged to be data entry errors, explained failures, or extreme failures are excluded from the data. A listing of these points is provided in the data anomaly section of the monthly IPR.

Where low and high standards have been identified for an instrument, the data is further separated into these categories. The mean (AVG %RDIF) and standard deviation (STD DEV %RDIF) of the weekly/monthly %RDIF data are computed and plotted on the charts above the number of the week of the calendar year. The accuracy plot displays the averages and the precision plot displays the standard deviations.

The symbols that are plotted on the x-axis of the accuracy plot indicate the occurrence of activities such as calibrations, repairs, and background corrections. Details on these activities can be found in the IPR. The IPR also includes a statistical and practical evaluation of each instrument's performance. In the literature on statistical process control there are numerous tests for non-randomness of the plotted data, including tests for trends and tests for bias. These tests provide "signals" that need to be evaluated. The Statistics Group reports on these signals, but also evaluates the data based on administrative limits. These limits, set by the operating groups and the Laboratory audit groups, are intended to help identify which signals of non-randomness have practical significance in the various processes. Thus, there may be a real bias present in a particular instrument, but that bias is so small relative to process and safeguards needs that it may be safely ignored. This will be demonstrated in the examples that follow.

Let us now consider some actual measurement control plots. The numbers printed below the week/month index number on the x-axis reflect the number of accuracy tests for each time period. When there are two rows of numbers, the upper number gives the number of low standard tests and the lower number gives the number of high standard tests.

Summary statistics are reported below the charts. The following terminology[d] is used.

STANDARD The value(s) of the standards. Because the standards used in the high and low categories may and do change, consider these values as representative rather than unique.

SYMBOL Solid or dashed line used to represent the data.

NUMBER WEEKS/MONTHS The number of weekly (monthly) periods for which there was data.

N The total number of accuracy tests considered after exclusion of anomalous data.

MEAN The mean of the N individual %RDIF values. These values are equal to the weighted average of the plotted AVG %RDIF values.

STANDARD DEVIATION

DAILY The standard deviation of the N individual %RDIF values.

POOLED[e] The pooled standard deviation of the grouped data.

SCALING This is a reminder that the plot scales may vary. The last line indicates how many (if any) accuracy test results were omitted.

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[c] For most instruments an adequate number of accuracy tests are performed weekly so that basic statistics may be estimated on a weekly basis. For some instruments, such as calorimeters, only one or two accuracy tests are performed weekly and hence the data is grouped by months for analysis. Note that we collected data by calendar week (Sunday–Saturday), so a year usually has data for 53 weeks.

[d] The terminology is changed appropriately for the electronic balances.

[e] The term "pooled" refers to a method of averaging the individual standard deviations. The individual weekly values are essentially weighted according to how many data points are in each week and then averaged. (For statistical reasons, averages consider variances rather than standard deviations.)

Figure 1 displays accuracy test statistics for a year on electronic balance B90. Because this is a balance, the accuracy tests are always performed in pairs, one for a low standard and one for a high standard. The standard values are approximately 1 kg and 4 kg. Accuracy tests were performed for 51 weeks (the Laboratory is closed during the Christmas/New Year's week), for a total of 166 accuracy tests. In the box at the lower right we see that no accuracy tests were excluded. The single row of numbers between the accuracy plot and the precision plot give the actual number of accuracy tests performed. Thus, in the first week four accuracy tests were performed and in the second week three were performed on each standard.

Over the 166 tests, the average deviation of the measured results from the standard value was 0.007 g for the low standard and –0.057 g for the high standard. These values are under the heading GRAND MEAN. The accuracy plot shows that the low standard weekly results were centered about the centerline, but that the high standard weekly results usually were a bit low and changed levels on occasion. The deviations shown by the high standard are statistically significant at times but were within the allowable administrative limit. Note that a "bias" of –0.1 g in the measurement of a 4 kg item represents an "error" of approximately $0.1 * 100/4000$, or 0.0025%.

The "precision plot" shows that the within-week variations (as measured by the standard deviation) were rather stable for each standard. The pooled weekly standard deviations were 0.039 g for the low standard and 0.058 g for the high standard.

Another summary statistic, the daily data standard deviation, is provided under the heading STD DEVIATION. For the low standard, the standard deviation of the 166 individual DIF values was 0.045 g; while for the high standard, the standard deviation of the 166 individual DIF values was 0.096 g. It is immediately seen that the daily data and the pooled weekly standard deviations are about the same for the low standard, but that the values are somewhat different for the high standard. This difference occurs because of the changes in level seen in the accuracy plot. A comparison of these two values is useful in assessing whether or not the instrument response is stable throughout the year. If it is, then the variation within a week should be about the same as the variation throughout the year and the daily data and pooled weekly standard deviations should be about the same.

Figure 2 displays a year of accuracy test data for Pu-239 on gamma counter G04. The standard values are approximately 9.6 g and 96.3 g of Pu-239. Unlike the electronic balances, accuracy tests are not done in low–high standard pairs, but individually on the low and high standards. So while there were 126 accuracy tests performed on each standard, the low standard checks covered 50 weeks, while the high standard checks covered 52 weeks. In the box at the lower right we see that no accuracy tests were excluded. The double rows of numbers between the accuracy plot and the precision plot give the actual number of accuracy tests performed on each standard. Thus, in the first week there were three accuracy tests performed on the low standard and two on the high standard.

Over the 126 tests, the average percent relative difference of the measured results from the standard value was 0.545% for the low standard and 0.322% for the high standard. These values are given under the heading MEAN (%RDIF). The accuracy plot shows that the standard weekly results were centered about the centerline for the first several months, but a bit high for most of the rest of the year for both low and high standards. Both biases were statistically significant, but considering the administrative limits for this instrument and the material being assayed, the biases were not deemed of practical importance.

The precision plot shows that the within-week variations (measured by the standard deviation) were rather stable for each standard. The pooled weekly standard deviations were 1.643% for the low standard and 1.804% for the high standard (Recall that the basic data considered is %RDIF, so the unit of the mean and standard deviation is %). The daily data standard deviations are just a bit higher than the pooled weekly. This is probably due to the apparent upward shift in the data.

Figure 3 displays a year of accuracy test data for Pu-239 on gamma counter G05. The standard values are approximately 9.6 g and 96.3 g of Pu-239 There were 198 accuracy tests performed on the low standard and 102 on the high standard. No accuracy tests were excluded. In the first week there were four accuracy tests performed on the low standard and one on the high standard.

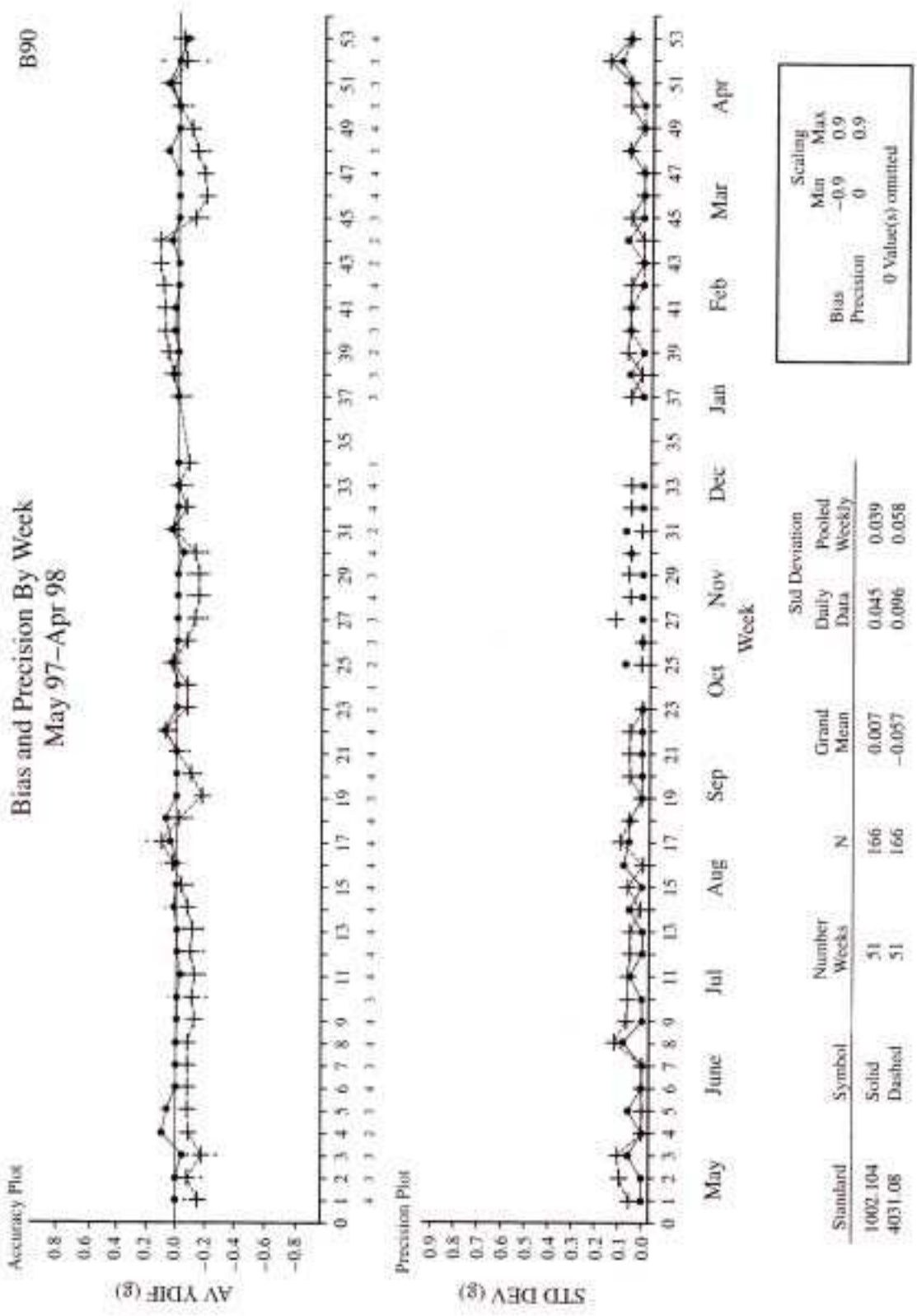

Fig. 1. Measurement control chart for a balance (B90).

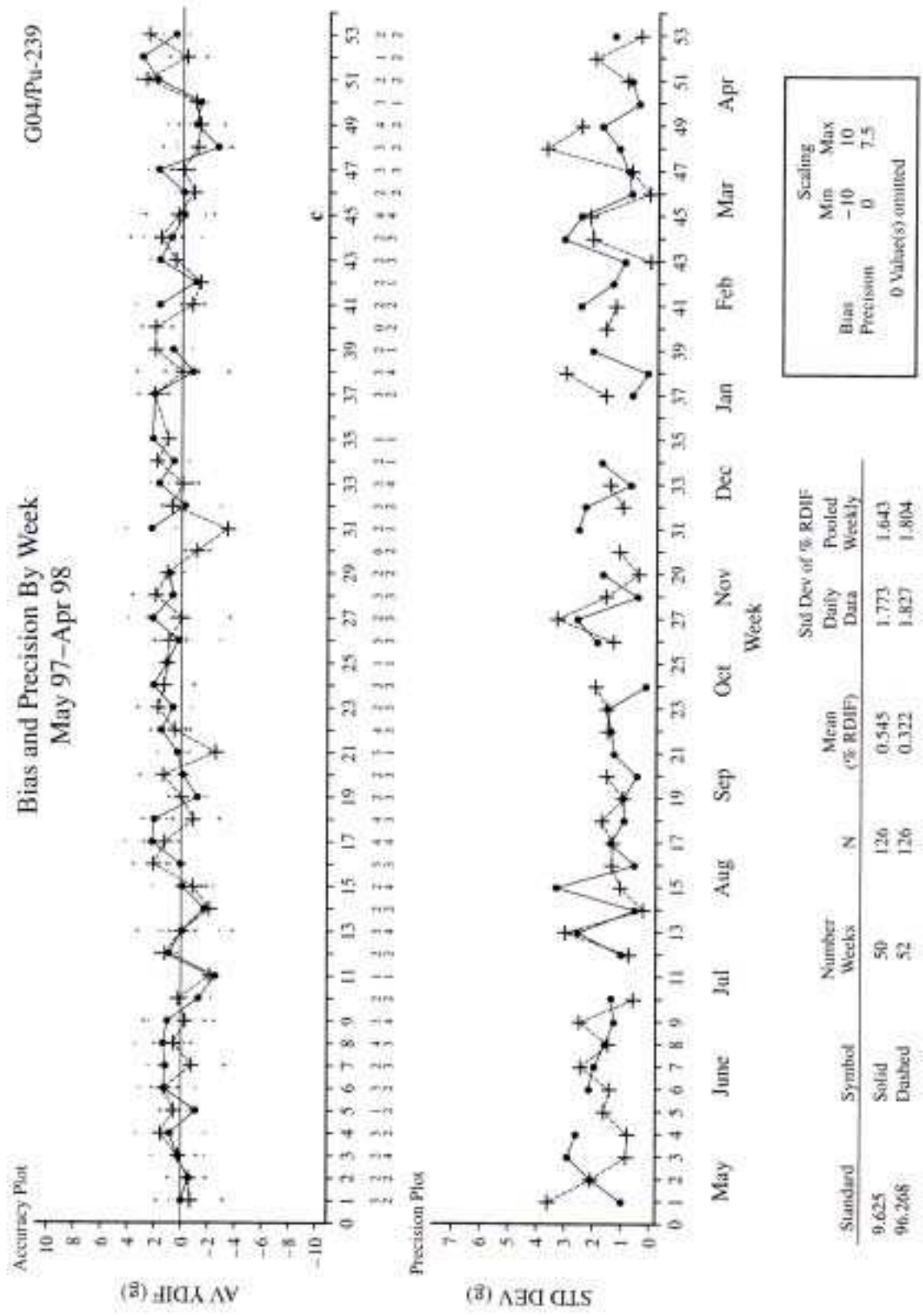

Fig. 2. Measurement control chart for a gamma counter (G04).

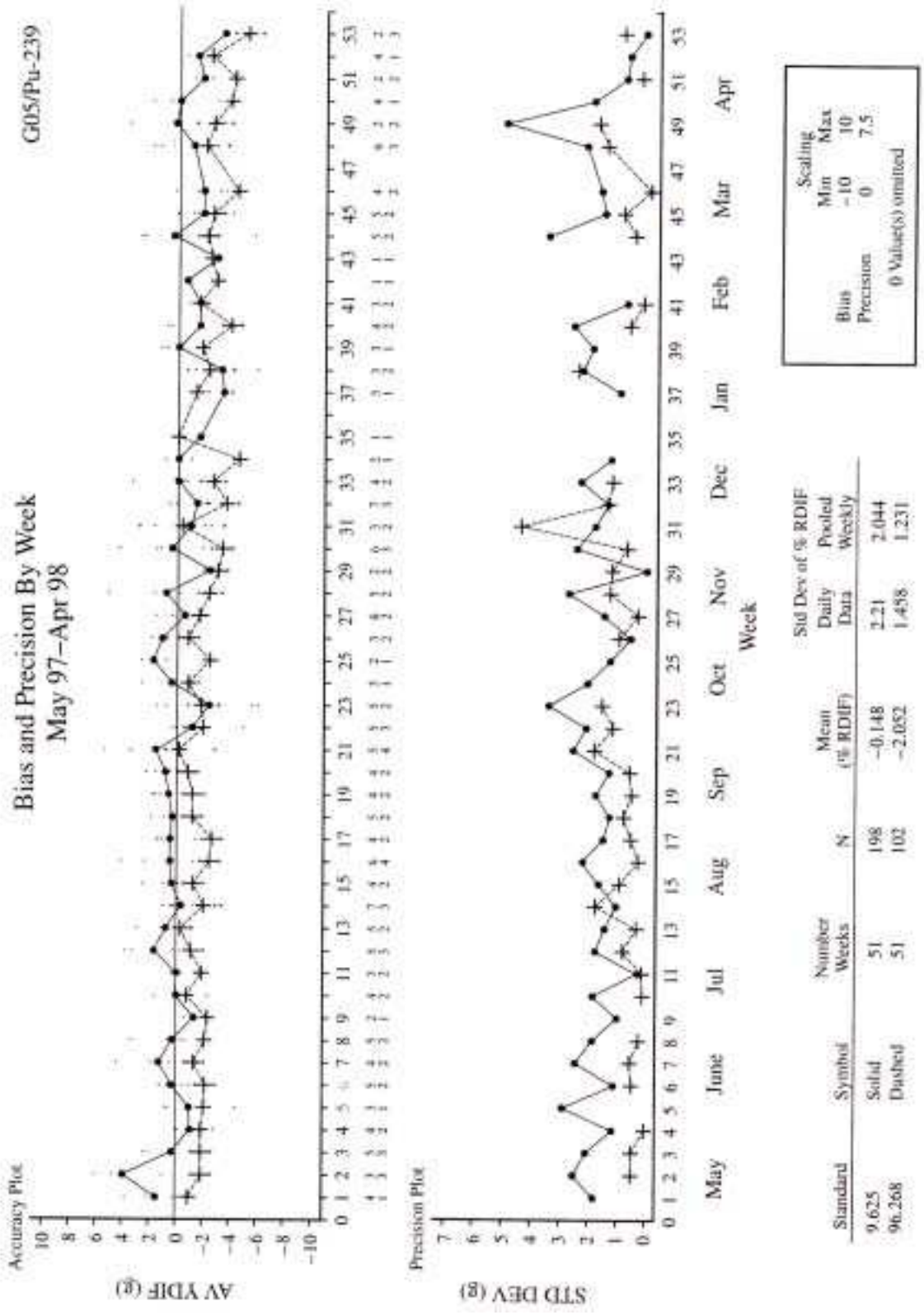

Fig. 3. Measurement control chart for a second gamma counter (G05).

The average percent relative difference of the measured results from the standard value was –0.148% for the low standard and –2.052% for the high standard. These values are under the heading Mean (%RDIF). The accuracy plot shows that the low standard weekly results were centered about the centerline for the first several months, but low for most of the rest of the year. The high standard ran low for the entire year. The plot indicates that about September/October the instrument response dropped for both standards. While the responses were within administrative limits for the entire period, the drop in week 53 resulted in a recalibration of the instrument.

The precision plot shows that the within-week variations were rather stable for each standard. There were high blips in November and in April that were investigated. The pooled weekly standard deviations were 2.044% for the low standard and 1.231% for the high standard. The daily data standard deviations are just a bit higher than the pooled weekly standard deviations, which is due to the downward shift in the data.

Figure 4 displays a year of accuracy test data for Pu-239 on solution assay instrument I08. The standard values are approximately 10.7 g Pu-239/L and 212.9 g Pu-239/L. The accuracy tests are performed individually on the low and high standards. There were 82 accuracy tests performed on the low standard and 79 on the high standard. No accuracy tests were excluded. In the first week there were four accuracy tests, one on the low standard and three on the high standard.

The average percent relative difference of the measured results from the standard value was 1.241% for the low standard and –0.657% for the high standard. The accuracy plot shows that the low standard weekly results were high for most of the year. The high standard ran low for most of the year.[f] There was a dramatic upward shift in the data in October, which may have been due to a recalibration of the instrument (Unfortunately, calibrations are not always reported to the Statistics Group for inclusion on the plot). Normally, one would expect to see the symbol ‘C’ on the plot at the date of calibration. The responses were within administrative limits for the entire period.

The precision plot shows that the within-week variations were rather stable for each standard. There was a high blip in April that was investigated. The pooled weekly standard deviations were 0.894% for the low standard and 0.756% for the high standard. The daily data standard deviation for the high standard is higher, by almost 0.3%, than the pooled weekly value. This is due to the change in data level in October.

VIII. FREQUENCY OF MEASUREMENT CONTROL TESTS

How often should you perform a measurement control test?[6,7] This depends on at least three primary factors:

- the instrument,
- the instrument’s working environment, and
- the consequences of a failure.

Obviously, the more stable and robust an instrument the less frequently you need to test to assure that the instrument is still in statistical control. However, this is not the only, nor necessarily the most important factor. The operating environment can dramatically impact even a stable, robust instrument. A poor power source, high humidity, rough treatment are only a few examples of environmental factors that can cause an instrument to go out of statistical control.

It is very important to consider a practical response to a measurement control failure. In section III of this paper, element number nine states, “After a measurement control failure, all items measured since the last passing measurement control are suspect items.” The question each operator needs to ask is, “Are there operational considerations that would limit my ability to measure suspect items following a measurement control failure?” For example, will all the items since the last passing measurement control test still be available considering your measurement control frequency? Even if all items are available, does your operational environment give you sufficient time to recover from a worst-case condition, namely, the remeasurement of all suspect items?

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[f] It is not uncommon to see a situation where one standard consistently runs high and another standard runs low. One explanation of this can be found in imperfect calibrations. Calibration curves are basically least-squares fits based on assumed models (for example, a linear fit through the origin). Because of variation present at the time of the calibration or a model that is not exact, the resulting calibration and quality control checks may exhibit the high/low patterns.

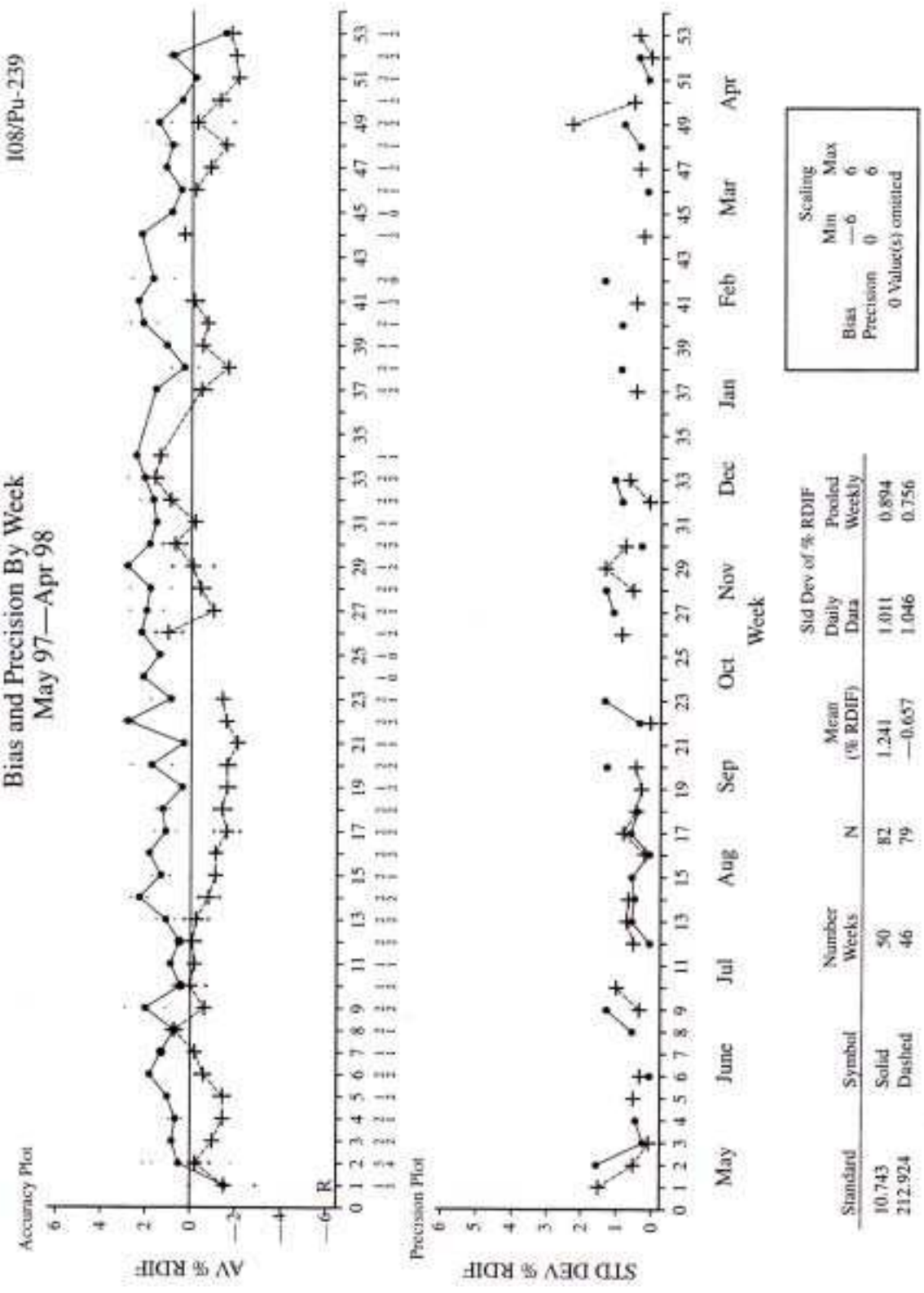

Fig. 4. Measurement control chart for solution assay instrument I08.

In general, for NDA instruments the recommended measurement control frequency would bound all measurements conducted during a single work shift. At our facility all of the gamma and neutron accountability instruments have a measurement control standard test done at the start and end of our day shift. However, the measurement control frequency is also driven by the assay time for the instrument.

In the case of calorimetry, we perform measurement control once per week. The typical measurement time for calorimeters varies by the size of the sample well and the heat-generating capacity of the item. For our calorimeters the range in measurement time is usually between 4 to 12 hours. It is clear that meeting the frequency of twice per day would mean only measurement control tests. Fortunately, calorimeters are extremely stable instruments. From our historical measurement control database, we have been able to demonstrate that they are stable to the point of having a warning and alarm failure rate well below that expected.

Even for instruments with short measurement times, we do not always bound all measurements with measurement control checks. Both our electronic balances and in-line NDA instruments have a measurement control test once per day in the morning. Electronic balances are extremely stable instruments. At our facility we have noted that they tend to fail catastrophically; that is, they either work perfectly or not at all. This property helps prevent erroneous data collection. There is another practical consideration in regard to measurement frequency on our electronic balances. We have over 100 such instruments at the Plutonium Facility. It is an intense effort to manually bring up so many instruments every morning to allow processing. That burden, in addition to lost productivity, would be exacerbated should we also have to perform another measurement control test at the end of every day. Again, the instrument's stability has spared us that necessity.

In addition to our NDA laboratory instruments, we also have gamma and neutron instruments in-line in the gloveboxes. This is a very harsh environment for the instruments compared with those in our laboratory. That alone would raise the argument that they should have measurement control tests at a higher frequency. However, we have chosen to define the measurements made on these instruments as temporary safeguard process control measurements. This is in contrast to our NDA laboratory that makes the final safeguards accountability measurements. With this interim status for our in-line instruments, we are willing to accept the associated risk of going out of statistical control because follow-on measurements will capture gross measurement errors associated with items leaving the process.

These follow-on measurements are assured by adhering to the approved process accountability flow diagram (PAFD) and daily ID analysis. The PAFD diagrams identify all of the process flows for nuclear material in a specific process and the required nuclear material measurements for each stream. An example is shown in Figure 5. This PAFD is a simplified copy of the cascade dissolver flow used to recover plutonium from our waste streams. Besides the flow diagram of the process, each input and output stream has circular symbols representing the types of measurement systems that must be used with these different matrices (see section X for elaboration on instrument names). No process can operate without such a PAFD. In addition, the daily ID analysis per PAFD assures that significant gains or losses are quickly identified and resolved.

Once the frequency has been defined, the time requirement is entered into the MCP frequency field. From this point the computer monitors and checks compliance with the required frequency. If the required frequency is not met, the computer system will not allow coding into our accountability system. This is discussed further in the next section.

The instruments in the Plutonium Facility have the following measurement control test frequency:

- balances—once per day,
- calorimeters—once per week,
- in-line NDA (gamma and neutron in process gloveboxes)—once per day, and
- off-line NDA (gamma and neutron in NDA laboratory)—twice per day, first and last measurement of the day.

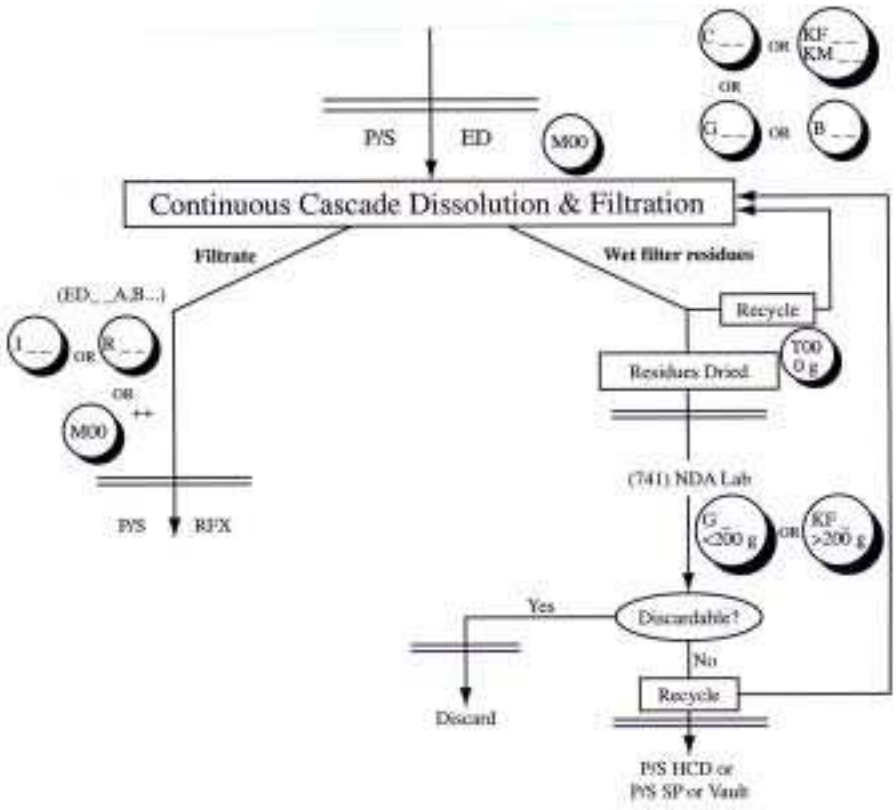

Figure 5. Cascade dissolver flow.

IX. TIE TO ACCOUNTABILITY SYSTEM

Element two from section III, states, "The MCP is tied into the safeguards accountability system to prevent data entry from instruments not currently passing the MCP." Our accountability system is the final authority on the location, type, and quantity of nuclear material present at our facility. It is this system that we, and any auditors, use as a basis for the facility's book value of all of our nuclear materials holdings. We have already stated the potential severe consequences if unquantified NDA biases exist on safeguarded nuclear material. This also holds true for errors in the accountability system that could be interpreted as an indication of potential diversion or theft. This risk is what has driven us to tie the MCP into the accountability system.

In order to enter the results from any NDA instrument into the accountability system, the MCP must indicate that the instrument currently has passed a measurement control test within the required frequency. So, if the NDA instrument has either failed its latest measurement control test or the operator has failed to conduct a measurement control test within the required frequency, the computer system prevents accounting data entry from this instrument into the accountability database. The current MCP system also allows management to manually place any system out of service to prevent data entry.

X. MANAGEMENT STRUCTURE

On the most basic level, any management structure that works to meet the facility's needs is acceptable. However, to mitigate the potential impacts from human error, a disgruntled employee, or safeguards all lead one to consider some level of "separation of duties" to assure adequate checks and balances. Separation of duties considers the use of additional layers of people or an intelligence-based computer system to either check the work of others, or partition the work so that a single individual does not have overall control.

The LANL Plutonium Facility is responsible for all instruments and all accountability and measurement control data entry. The different tasks associated with this are split amongst different teams. For example, our NDA analytical laboratory performs all of the measurement control measurements and product measurements on all off-line instruments and enters the measurement control results into the MCP system. They do not enter product measurement results into the accountability system. This is reserved for the operators who requested the product measurements, or our accountability office for compliance-based measurements (typically based on a DOE safeguards request or DOE requirement). A separate team performs the calibration and maintenance on this equipment. For in-line instruments, the arrangement is different. The operators with instruments in their gloveboxes are responsible for all product measurements and entering the data into the accountability system. Calibration, maintenance, and all measurement control measurements and data entry into the MCP on this equipment is performed by the same team that maintains the off-line instruments.

An independent group is responsible for maintaining the data portion of the computer-based MCP. This includes all records concerning instrument limits, measurement ranges, standard values, measurement control test frequency, and placing instruments in and out of service. An independent statistician supports the data portions of the MCP system. This support includes evaluating certification data on new instruments and historical data on existing instruments. Currently, all limits are evaluated annually. The independent group also supports development and evaluation of measurement control algorithms. An independent computer team supports the computer hardware and software.

XI. ELEMENTS OF THE SYSTEM

One of the most important practical aspects of any MCP system is the ease of data entry for operators. It is imperative to have either readily accessible data entry devices such as local plots to graph new points, computer terminals, barcode readers, or direct data transfer through electronic connections to computer terminals/PCs.

The following items are the primary elements of our MCP system.

- Unique instrument name for each instrument. For example, our current system is limited to just four characters in what we refer to as the measurement code field. As a result of this restriction the following are some LANL codes and their meanings.

  G02—segmented gamma scanner number two (the second such system entered into the MCP)

  N01—passive thermal neutron coincidence counter one (the first such system entered into the MCP)

  B125—electronic balance (125th entered into the system; we reserve a sequence of numbers for a specific facility when appropriate)

  K1—calorimeter number one (specifically limited to a two character description because of the unique requirement to be associated with a functional gamma isotopic unit that provides quantitative measurement)

  F5—gamma isotopic unit number five (specifically limited to a two character description because of the unique requirement to be associated with a functional calorimeter that provides quantitative measurement)

In the case for calorimetry, the MCP must assure that two instruments, namely a calorimeter and an isotopic unit, have passed measurement control to allow coding on the accountability system.

- Instrument-specific uncertainty limits. This includes a 1 sigma limit used for both the warning and action calculations and an administrative limit if required.
- Unique standard name for each standard on the MCP.
- Value of standards. These standards must be certified with a certificate indicating the quantity, basis of certified value, and uncertainty for each stated isotope. Separate computer fields are provided to enter the value of each isotope.
- Partitioned computer access for facility operator and independent group. As part of the separation of duties requirement for safeguards (see section IX), the MCP menu for activities has been partitioned to restrict access based on an individual's job assignment.

For example, individuals who perform NDA measurements are restricted to 3 of 10 access levels. One gives the status of the instrument in regard to its availability for measurements. The second allows entry of measurement control data for a specific instrument. The third allows one to view the last three months of MCP test data.

Others who monitor the MCP are restricted from entering the data. They have access to the edit functions of the MCP which allow modifications to instrument MCP limits, standard limits, and the frequency of MCP tests.

XII. HOW IT WORKS: DAY TO DAY AND MONTH TO MONTH

Day to Day

A measurement control standard is measured on the required instrument at the required frequency dictated by the MCP. This is routinely done at the start and end of a workday for those instruments requiring daily measurement control tests and producing final accountability values (see section VII).

An instrument operator manually enters data into the measurement control computer system. The MCP has a built-in check before accepting the data. It displays the data entered and asks if this entry is correct. A second “yes” entry actually sends the data to the MCP.

After confirmation of the data entry, the MCP computer uses the computer-based standard value and the instrument uncertainty limits, to compare the entry with the measured value. Then the computer indicates either a pass or the type of failure with the following actions to be taken by the operator.

- If pass, operator continues with assays until next measurement control standard must be measured.
- If warning failure, standard measurement is repeated.
- If pass, continue with assays.
- If this is a second warning failure (equivalent to an action failure) or any action failure, computer places the instrument out of service.

Whenever an instrument is placed out of service, either by the MCP or by the operator, an investigation into the cause of the failure or degraded performance is conducted. All investigative actions are documented locally by the NDA lab operator. This is typically captured in the instrument log (either computer-based or in a logbook).

Upon completion of the investigation, a report stays in the NDA lab file, a file number is assigned to this investigation, and it is referred to the independent group for its failure report, a short summary of the actions taken and references the identified failure indication on the computer database. This completes the linkage between the instrument logbook and the computer database failure record, thereby assuring traceablity.

The instrument is now ready to be placed back in service. To accomplish this, the instrument operator runs a measurement control test and enters the data on the MCP. Assuming that this test passes, the operator notifies the independent group which reviews the MCP data, confirms that it has passed, and places the instrument back in service.

Month to Month

At the end of each month, the computer database operators transfer the MCP data to the Statistics Group (see Table 2). The statistician generates the MCP charts for each instrument. A review is made of each measurement control plot (see section VII). The statistician compiles the review table (see Table 3). This table indicates whether the instruments met the desired levels of precision and bias. A comment field allows the statistician to point out other items of interest such as potential trends or possible biases.

Table 2. Measurement Control History Repoirt

Date	Time	TT	OP	Standard	Mass	Meas	Diff	Accy	Standard	Mass	Meas	P/F	Diff	Accy
951117	11:49	3	JEM	MD3	500.048	500.050	0.002	0.036	39A	4000.003	3999.980		–0.023	–0.403
951128	10:00	3	MP	MD3	500.048	500.070	0.002	0.388	39A	4000.003	3999.960		–0.043	–0.754
951129	10:02	3	MP	MD3	500.048	500.060	0.012	0.212	39A	4000.003	3999.990		–0.013	–0.223
951130	10:52	3	MP	MD3	500.048	500.040	–0.008	–0.139	39A	4000.003	4000.010		0.007	0.120
B134														
951101	10:09	3	MP	71A	10.000	10.002	0.002	0.272	71B	100.000	100.001		0.001	0.120
951107	9:50	3	CAB	71A	10.000	9.999	–0.001	–0.157	71B	100.000	100.001		0.001	0.120
951108	9:06	3	MP	71A	10.000	10.000	0.000	–0.014	71B	100.000	100.000		0.000	–0.024
951114	11:26	3	JEM	71A	10.000	10.000	0.000	–0.014	71B	100.000	99.999		–0.001	–0.168
951115	13:36	3	ALH	71A	10.000	10.001	0.001	0.129	71B	100.000	100.000		0.000	–0.024
951116	11:05	3	ALH	71A	10.000	10.000	0.000	–0.014	71B	100.000	100.000		0.000	–0.024
951117	11:50	3	JEM	71A	10.000	10.001	0.001	0.129	71B	100.000	100.001		0.001	0.120
951120	14:05	3	MP	71A	10.000	10.001	0.001	0.129	71B	100.000	100.000		0.000	–0.024
951129	10:02	3	MP	71A	10.000	10.002	0.002	0.272	71B	100.000	100.001		0.001	0.120
951130	10:52	3	MP	71A	10.000	10.001	0.001	0.129	71B	100.000	100.000		0.000	–0.024
B136														
951101	10:16	3	MP	3Q	100.000	100.000	0.000	–0.003	3C	1005.395	1005.370		–0.025	–0.281
951103	9:55	3	JEM	3Q	100.000	100.000	0.000	–0.003	3C	1005.395	1005.370		–0.025	–0.281
951106	9:20	3	MP	3Q	100.000	100.000	0.000	–0.003	3C	1005.395	1005.370		–0.025	–0.281
951107	15:34	3	JEM	3Q	100.000	100.000	0.000	–0.003	3C	1005.395	1005.370		–0.025	–0.281
951108	9:16	3	MP	3Q	100.000	100.000	0.000	–0.003	3C	1005.395	1005.370		–0.025	–0.281
951114	11:27	3	JEM	3Q	100.000	100.000	0.000	–0.003	3C	1005.395	1005.370		–0.025	–0.281
951115	15:50	3	ALH	3Q	100.000	100.000	0.000	–0.003	3C	1005.395	1005.370		–0.025	–0.281
951116	10:35	3	ALH	3Q	100.000	100.000	0.000	–0.003	3C	1005.395	1005.370		–0.025	–0.281
951117	11:53	3	JEM	3Q	100.000	100.000	0.000	–0.003	3C	1005.395	1005.370		–0.025	–0.281
951120	13:58	3	MP	3Q	100.000	100.000	0.000	–0.003	3C	1005.395	1005.370		–0.015	–0.169
951128	10:04	3	MP	3Q	100.000	100.000	0.000	–0.003	3C	1005.395	1005.370		–0.035	–0.394
951130	10:57	3	MP	3Q	100.000	100.010	0.010	0.109	3C	1005.395	1005.370		–0.025	–0.281
B137														
951101	10:10	3	MP	MD2	500.036	500.030	–0.006	–0.100	41A	4000.000	4000.030		0.030	0.496
951107	9:52	3	CAB	MD2	500.036	500.050	0.014	0.233	41A	4000.000	4000.060		0.060	1.001
951108	9:09	3	MP	MD2	500.036	500.050	0.014	0.233	41A	4000.000	4000.050		0.050	0.830
951115	13:38	3	ALH	MD2	500.036	500.050	0.014	0.233	41A	4000.000	4000.050		0.050	0.830
951117	8:42	3	ALH	MD2	500.036	500.050	0.014	0.233	41A	4000.000	4000.050		0.050	0.830
951117	9:13	3	RJM	MD2	500.036	500.050	0.014	0.233	41A	4000.000	4000.050		0.050	0.830
B142														
951101	8:59	3	JEM	3N	400.002	400.000	–0.002	–0.177	3H	4041.390	4041.370		–0.020	–2.002
951103	10:05	3	JEM	3N	400.002	399.990	–0.012	–0.178	3H	4041.390	4041.360		–0.030	–3.027
951106	8:37	3	JEM	3N	400.002	400.000	–0.002	–0.177	3H	4041.390	4041.370		–0.020	–2.002
951107	9:43	3	JEM	3N	400.002	400.010	0.008	0.824	3H	4041.390	4041.570	FN	0.180	17.969
951107	9:43	3	JEM	CAL	1.000	1.000	0.000	0.000	CAL	1.000	1.000		0.000	0.000
951107	9:44	3	JEM	3N	400.002	400.000	–0.002	–0.177	3H	4041.390	4041.390		0.000	0.000
951108	9:58	3	JEM	3N	400.002	399.990	–0.012	–0.178	3H	4041.390	4041.380		–0.010	–1.025
951114	8:23	3	JEM	3N	400.002	399.990	–0.012	–0.178	3H	4041.390	4041.390		0.000	0.000
951115	7:54	3	JEM	3N	400.002	399.990	–0.012	–0.178	3H	4041.390	4041.390		0.000	0.000
951116	9:10	3	JEM	3N	400.002	400.000	–0.002	–0.177	3H	4041.390	4041.410		0.020	2.002

Table 3. Statistician's Review

Code	Desired Levels of Bias	Precision	Meets Desired Level ❑ = Y	Comments	Discuss
K8-low	0.2%				
-high				No new data	
K9	0.5%			Certified but not used	
KA-low	0.2%			Bias continuing at about 0.2% low	
-high				Bias continuing at about 0.2% low	
KE-low	0.3%			Bias continuing at about 0.2% low	
-high				Bias continuing at about 0.2% low	
KF-elect	0.2%			No new data	
-rad					
KTWI	0.2%			Increasing positive bias over last two months. About 0.7%, but only two data points	
BD1	0.8%			No new data	
BD2					
G03-238				Two failures with no reports	
-239	2%				
-U				No new data	

This data is reviewed at a monthly or quarterly measurement control meeting attended by the instrument operators (for those instruments requiring a review, noted on the statistician's review table), oversight organization, and statistician. A consensus is reached on any actions, including suspect trends. This might result in a decision to calibrate an instrument or to track the potential trend. Others decisions may be made. For example, if changes to the MCP algorithms or new algorithms are to be discussed, then the computer database operators will also be asked to attend. In any case, it is intended that all MCP issues be resolved at this level. When this is not possible, issues are elevated to the next level of management.

An annual review is made of all limits to determine if any adjustments are warranted. This includes a review of failure rates and the acceptable levels of bias and precision.

XIII. COMMON CAUSES OF FAILURE

The current MCP system has been basically unchanged since the mid-1980s. We see common failure modes that impact our system, including the following.

1. Typographical errors—Over 90% of our measurement control failures are due to typographical errors. This is not surprising considering that the technicians must manually enter measurement control data on more than 100 balances and 40 other NDA instruments every workday morning.
2. Balances: moved in glovebox—Electronic balances are very robust instruments. However, if they are moved, the calibration is disturbed. The nature of activities in our gloveboxes necessitates moving our electronic balances to accommodate ongoing work.
3. Storage or movement of radioactive items close to a NDA instrument—NDA instrumentation was not considered in design of the LANL Plutonium Facility. As a result, nuclear material is routinely transported and stored adjacent to our instrumentation. Lack of space has also forced us to co-locate instruments in a close-packed arrangement. Lack of shielding makes the NDA measurements susceptible to interferences from nuclear material. Because of the

continuously changing nuclear material quantities, the radiation background is highly variable. There is both a reduction in sensitivity and increased uncertainty in the measured result.

4. Electrical fluctuations and outages—All of our instruments are on plant power. Electrical transients have the greatest impact on our NDA systems. We primarily see the impact to our computers and associated algorithms. This requires a complete system restart to clear the problem.
5. Contamination from the previous item in an in-line instrument—For our in-line TNCs and particularly our SAIs, contamination in the instrument's sample chamber can result in a potential high bias. The TNCs may be contaminated by oxide powders and the SAIs from solution that leaks from the sample containers.

XIV. SOME POSSIBLE FIXES

It is not difficult to identify potential upgrades that would help minimize or eliminate the problems identified in section XII.

1. Input data directly into the computer—Manual data entry is causing the majority of our false failures on the measurement control system. Direct data transfer from instruments to the measurement control system will eliminate this problem. A new integrated nuclear material management information system is being designed.
2. Locate NDA lab in low background area or shield instruments—The Plutonium Facility was not designed for NDA. Some instruments will have shielding installed. Because this was not incorporated in the original instrument design, this will be very limited. Space limitations make it difficult to shield many of these instruments without impacting required access for servicing. We are planning to redesign some instruments as part of an upgrade. In addition, we are exploring new locations for our waste instruments. Because of the low mass quantities we are striving to measure, these instruments are particularly susceptible to a highly variable radiation background.
3. Control item flow to minimize impact on instruments—Part of the investment to upgrade our instrumentation includes providing shielded staging/storage areas for our NDA labs. We are currently constructing carts that use both neutron and gamma shielding. This will not only help the instruments, but will help us reduce personnel exposures.
4. Provide "clean" power, independent from the remainder of the facility—Ideally, independent clean power would be preferred for our NDA instruments. There are plans for a facility-wide uninterruptable power supply (UPS) system. Until then, we are adding individual UPS systems to each PC on each instrument.
5. Better containers for solutions and algorithms to test for contamination—Solutions themselves are problematic. Poor filling techniques or pressurization due to radiolysis can cause contamination of the instrument's sample wells. This is also true of powders. Better-designed containers can prevent the current problems. Additionally, smart algorithms with well-shielded measurement wells can determine when elevated backgrounds are due to sample well contamination.

XV. SUMMARY

Our current measurement control program was first implemented in 1984. It is written in FORTRAN. A new system is under development to address multiple shortcomings. Nevertheless, the current system has proved to be very robust, both in terms of service to the facility and in response to internal and external audits. We have attempted to present all of the considerations employed to develop our current measurement control system, from the initial considerations for the system's design to the maintenance of an efficient functional system. This effort is the product of many individuals over the decade and a-half since it was first initiated. Besides the compliance-based reasons for such a system, it is the authors' hope that the reader gains insight into the consideration for such a system, and also an appreciation for the facility-wide impact such a system can have on both inventory and process knowledge.

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