MC&A IN A RADIOCHEMICAL PLANT

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Abstract

MC&A in reprocessing plant in the United States is based on solution measurements of the dissolved fuel assemblies, periodic inventories, and solution measurements of product and waste streams. At the Savannah River Site, SRS, considerable effort has been involved in obtaining accurate measurements of the input and product output streams including both volume analytical measurements. In general, volume measurements are based on differential pressure measurements of level and solution density. Topics being addressed are

- the calibration of tanks in a reprocessing plant,
- the instrumentation used for the input and product volume measurements,
- the analytical measurement capabilities used for input, output and waste streams,
- the measurement uncertainty of these measurements,
- the inventory measurement techniques used at SRS,
- the frequency of material balances,
- a summary of typical measurement system uncertainties, and
- typical material balance uncertainties achieved at SRS.

An overview of these topics with examples in each of these areas based on SRS experience will be provided.

Introduction

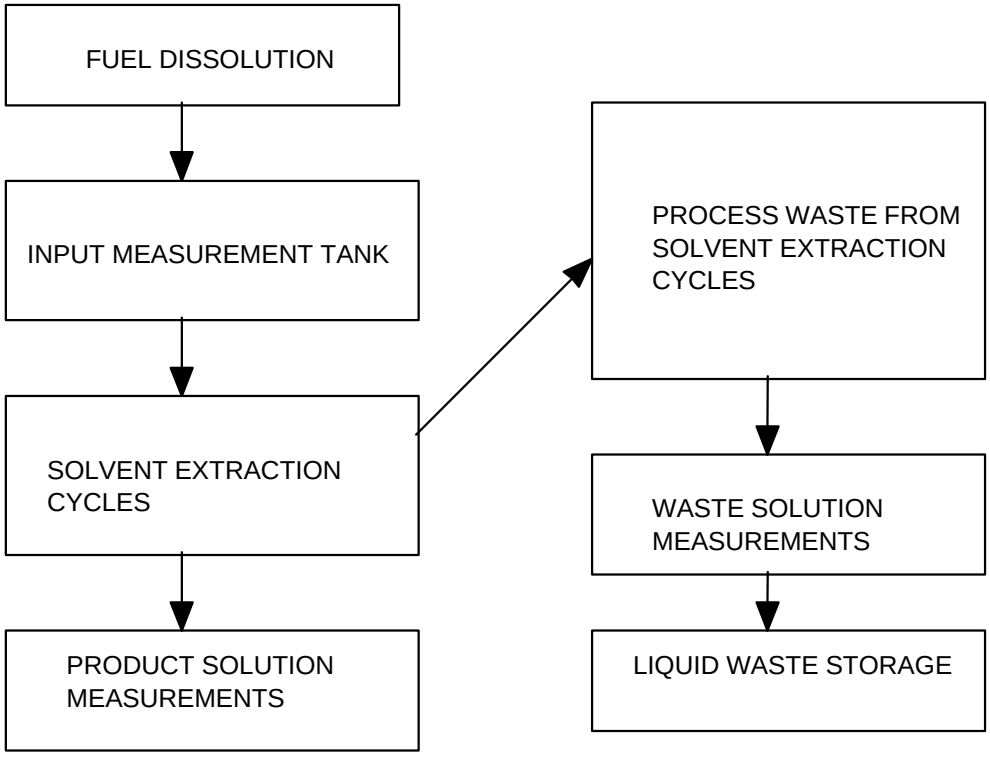
The Savannah River Site (SRS) was established in 1950. Its primary mission was to provide strategic isotopes (plutonium-239 and tritium) used in the development and production of nuclear weapons for national defense. The historical production cycle at SRS involved fabrication of metal fuel and target assemblies for irradiation in the site reactors, followed by chemical dissolution, separations, and conversion into solid forms. The current mission is to store, treat, stabilize, and dispose of waste material; manage nuclear materials and facilities; and restore and manage the site's natural resources.

The Savannah River Site is located in southwestern South Carolina. It occupies approximately 198,000 acres (310 square miles), and it is 25 miles from Augusta, Georgia. The site is composed of forest, undeveloped land, wetlands, streams, and lakes. The facilities at SRS include 16 major production, service, and research and development areas. Some of these facilities are not currently operating.

The separation process consists of dissolution of fuel, separation and purification of actinides to a nitrate product. The measurement systems in use at SRS to determine material balances around this process will be described. The measurement capabilities achieved in a production plant are not always the same as those achieved in a research or a laboratory environment. It is expected that SRS experience parallel much of the experiences in Russia. In general, the error estimates provided are obtained from measurement control programs in place at SRS to control, monitor and estimate measurement error and from tests performed to estimate measurement errors.

A simplified material balance flow chart for the separations material balance is provided as Figure 1.

FIGURE 1 SIMPLIFIED MC&A MATERIAL



Volume Measurements

The separation plants at SRS use bubbler probes to determine tank volumes. There is a long history of using this technique for measuring solutions in US separations (radiochemical) plants. Obviously, the quality of these measurements is dependent on the measuring instrument and the tank calibration. In the late 1970s to early 1980s, SRS started to use more accurate differential pressure instruments together with extensive tank calibrations to reduce the volume measurement error. Typical volume uncertainties were about 0.5 to 2 % for input and output product tanks before this change. Currently all of the input and product tanks in SRS separation plants use the more accurate differential pressure measurements and have detailed calibration equations. The tank volume errors are consistent with the maximum target error limits of uncertainty for primary accountability tanks of 0.3 % random and 0.2% systematic (IAEA/ANS/INMM-INMM 1994)[(1)]
less than 0.2% at one sigma for both random and systematic errors. For our best calibrations uncertainties of less than 0.1% at one sigma for both random and systematic errors were achieved.

uncertainties from 1 to 5 % random and systematic errors. The calibrations for these tanks are considered adequate based on the amount of material measured. The larger uncertainties do not significantly increase

To ensure that volume accuracy's are as expected, SRS has implemented a volumetric measurement control program to control errors and to detect anomalies. The program consists of periodic instrument calibrations,

and comparison of in-tank density measurements. These types of checks have proven beneficial in improving the reliability of volume measurements and in reducing the number of anomalies.

One of the separation plants at SRS has processed primarily Highly Enriched Uranium (HEU) while the other has primarily processed Plutonium and depleted Uranium. Therefore, analytical measurement

estimates provided are from analysis of current analytical measurement control data.
The uranium measurement methods in use at SRS are the standard methods in use in the U. S. These are

- Isotope dilution mass spec for input, impure inventory and waste solutions. The uncertainty of this
- potentiometric titration) method for lower activity uranium solution. This method is used for product solutions and has an uncertainty estimate of about 0.2 % random and 0.1%
- phosphorescence) for low concentration levels. The uncertainty of this method as used at SRS is about 10.0 % random and 6% systematic

The plutonium measurement methods in use at SRS are the standard methods in use in the US. These are

- Coulometry is used for product measurements and as the method of choice to characterize solutions for other methods. The uncertainty of this method as used at SRS is about 0.2 % random and 0.1 % systematic.
- Diode Array Spectrometer (DAS) is used to determine plutonium concentration in input solution as well as for many tanks on inventory. The uncertainty of this method as used at SRS ranges from 1 to 5 % random and 1 to 5 % systematic depending on the concentration and control used.

- Alpha counting techniques are used to determine plutonium concentration for waste output and inventory. This method includes several adaptations such as gross alpha without corrections, alpha count after removing impurities, or gross alpha with corrections for alpha activity from interfering isotopes (Am and Pu-238). The uncertainty of this method as used at SRS varies from 1 to 5 % random and 3 to 5 % systematic.

An analytical quality control program is in place at SRS to provide assurances that analyses are reliable and to estimate and monitor method performance. Checks include the analysis of standards prior to the use of each method each shift the method is used. The program uses standards that match the material being analyzed in both matrix and concentration.

As an additional control check on our analytical measurements, SRS participates in a sample exchange program administered by the New Brunswick Laboratory (NBL). Material is sub-sampled with these samples sent to different laboratories in the U.S. NBL collects the data, analyses the results, and issues reports on the performance of laboratories.

Inventory Measurements

Currently inventories are conducted every two months at SRS separation plants. The inventory steps are

- cutoff of material flows into and out of process,
- ensure that the mixer-settler systems are brought to standard or known condition for inventory,
- transfer and collect solution in measurable locations,
- mix and sample tanks,
- monitor sample quality from replicate samples,
- analyze and report sample results,
- calculate inventory quantity, and
- compare book value to inventory results

At SRS for several years, each inventory difference is compared to the combined errors for the balance. That is the error estimates for input, output including waste, and beginning and ending inventory are combined by variance propagation techniques to determine total uncertainty on the balance. Care must be taken to ensure that the measurement correlation is correctly included in the analysis. Any inventory difference that exceeds the calculated 99 % uncertainty limits is investigated. This is one of the primary ways used to judge the adequacy of the inventory.

Overall Balance

For many years the inventory differences were tracked and compared to historical limits. The results of some of these inventory differences are plotted in Figure 2 for High Enriched Uranium (HEU). The equivalent data for plutonium is provided in Figure 3. These show that there were long-term measurement biases in one or more of the measurement systems. These plots are for the entire site and reflect more than just the separation balances. However, separation processes were contributors to the cumulative balances. Trends are apparent in much of this historical data.

FIGURE 2

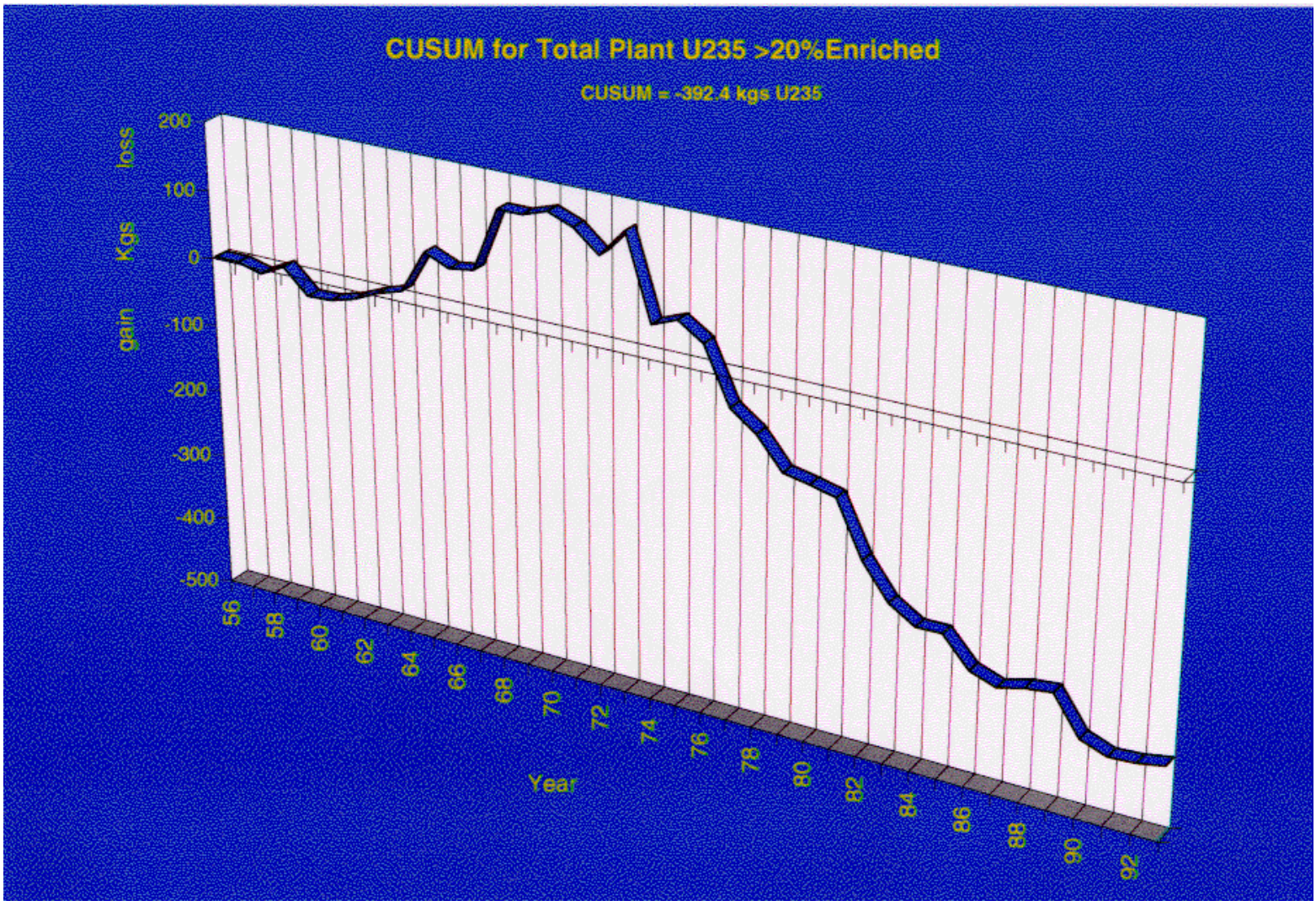
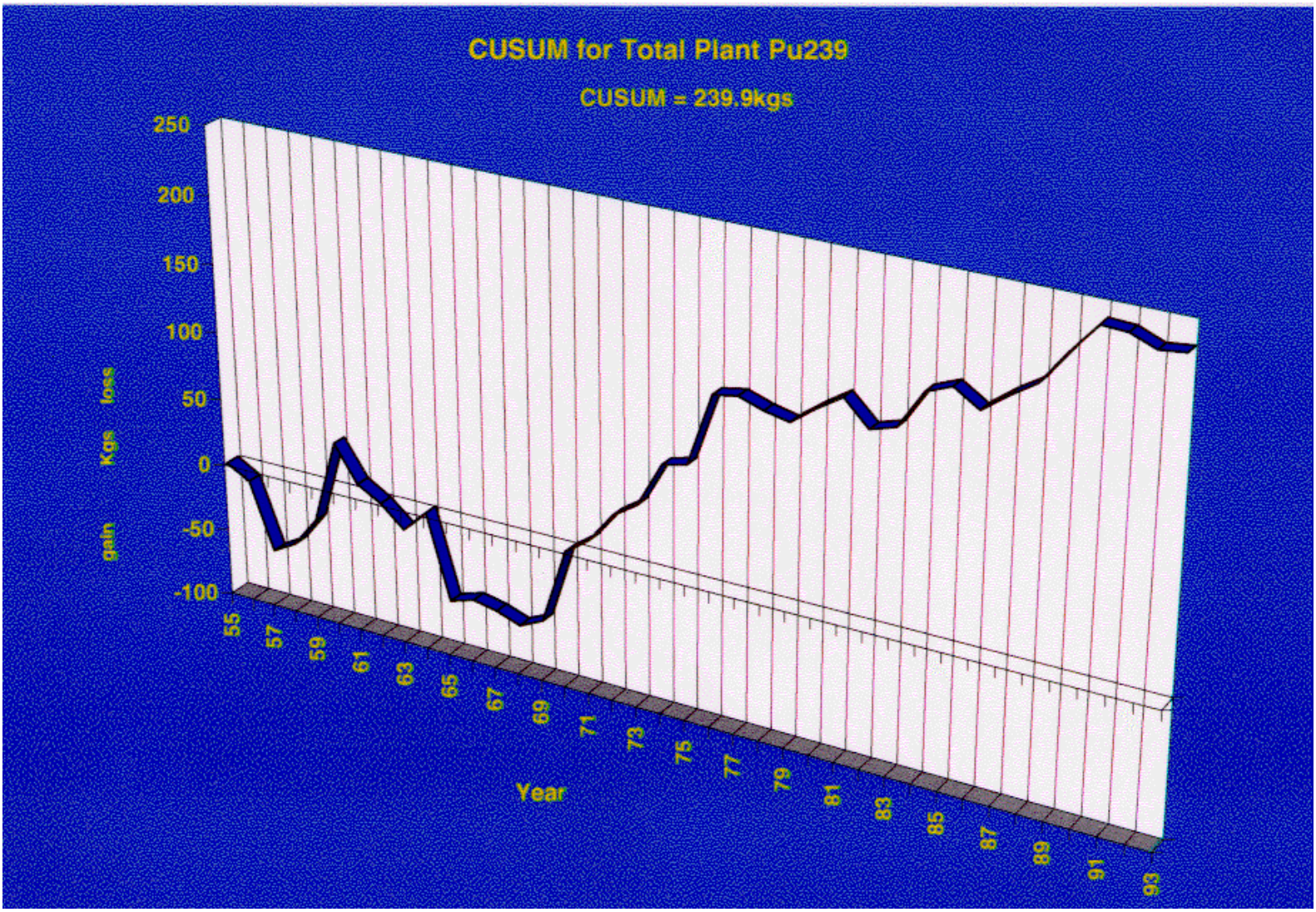

CUSUM for Total Plant U235 >20%Enriched
CUSUM = -392.4 kgs U235
gain kgs loss
200
100
0
-100
-200
-300
-400
-500
56
58
60
62
64
66
68
70
72
74
76
78
80
82
84
86
88
90
92
Year

FIGURE 3


CUSUM for Total Plant Pu239
CUSUM = 239.9kgs
250
200
150
100
50
0
-50
-100
gain Kgs loss
55
57
59
61
63
65
67
69
71
73
75
77
79
81
83
85
87
89
91
93
Year

At SRS for several years, cumulative inventory difference is compared to the combined errors for the balance. That is the error estimates for input, output including waste, and beginning and ending inventory are combined by variance propagation techniques to determine total uncertainty on the balance. Care must be taken to ensure that the measurement correlation is correctly included in the analysis. The results from current evaluations are presented in Figure 4 for uranium balances and in Figure 5 for plutonium balances. These are considered typical of performance for the last several years. This represents the magnitude of errors experienced at SRS and what can be achieved from propagating the measurement errors described in the above paragraphs.

To put these data into a meaningful perspective, the cumulative balance data for HEU is about 2 % of the active inventory. These numbers are from a time period with relatively small throughput compared to the historical plots.

Similarly, the Pu cumulative balance data is based on relatively low throughput values with decreasing inventory values. These values vary between 2 and 3 % of the active inventory.

FIGURE 4

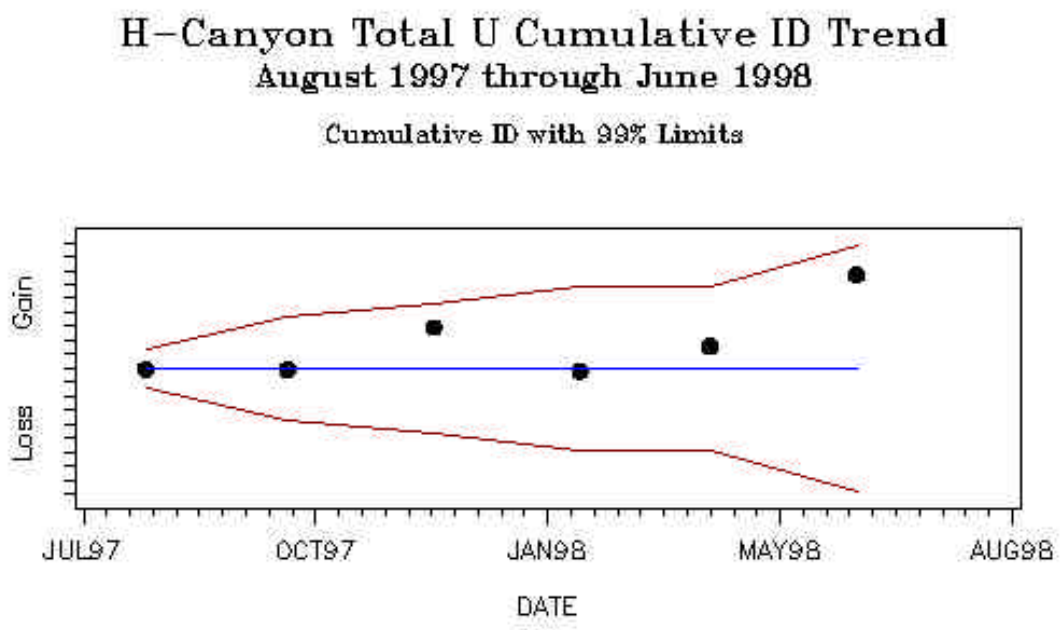
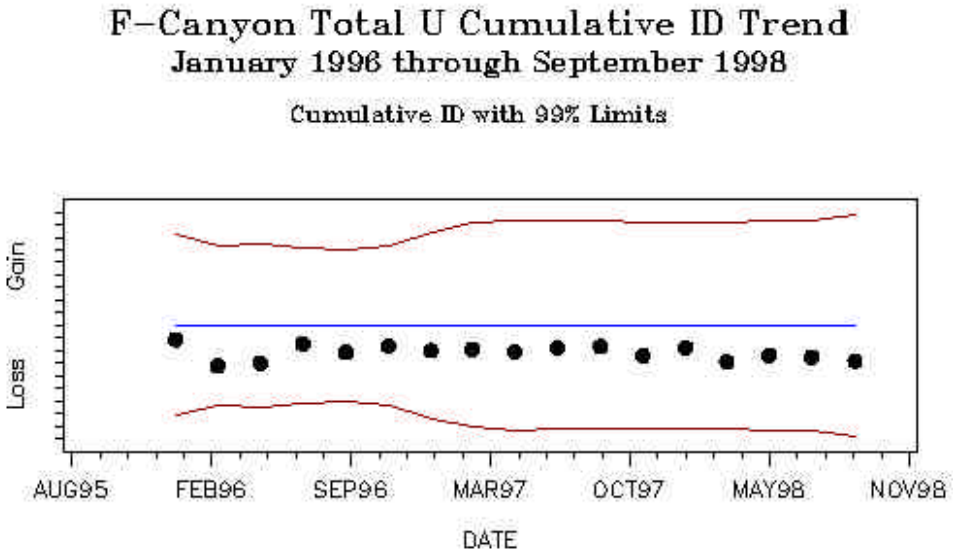


FIGURE 5



References

B. A. Hunt, et. al. , Mass/Volume Techniques and Methodologies Applied in Nuclear Facilities, Proceedings of the Tripartite Seminar on Nuclear Material Accounting & Control at Fuel Fabrication Plant, April 21-26, 1997 at Obninsk, Russia