Using Statistical Process Control to Monitor Radioactive Waste Characterization at a Radioactive Facility

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ABSTRACT

Two facilities for storing spent nuclear fuel underwater at the Hanford site in southeastern Washington State are being removed from service, decommissioned, and prepared for eventual demolition. The fuel-storage facilities consist of two separate basins called K East (KE) and K West (KW) that are large subsurface concrete pools filled with water, with a containment structure over each. The basins presently contain sludge, debris, and equipment that have accumulated over the years.

The spent fuel has been removed from the basins. The process for removing the remaining sludge, equipment, and structure has been initiated for the basins. Ongoing removal operations generate solid waste that is being treated as required, and then disposed. The waste, equipment and building structures must be characterized to properly manage, ship, treat (if necessary), and dispose as radioactive waste. As the work progresses, it is expected that radiological conditions in each basin may change as radioactive materials are being moved within and between the basins. It is imperative that these changing conditions be monitored so that radioactive characterization of waste is adjusted as necessary.

At the beginning of the decommissioning activities, a method was chosen to monitor the radioactive characteristics of the basins by analyzing samples of basin water. However, it was quickly discovered that as fuel-removal activities changed, the fluctuations in water conditions were too severe to act as a useful indicator. Therefore, a different method for monitoring radioactive characteristics was required that would be more stable, and yet, sensitive to important changes in conditions. The routine survey data for controlling radiological surface contamination was selected. There were two reasons for this choice: the data is a direct measurement of the material being removed as waste; and this information was already being collected, and therefore did not require additional resources. Statistical process control was chosen as the method to use to evaluate the data, as it has long history of successful industrial use, identifies trends in a timely manner, is flexible, and is the simplest statistical analysis that separates signal from noise. The statistical process control method that was applied utilizes the industry practices of W. A. Shewhart and W. E. Deming.

To apply statistical process control, a baseline of radioactive characteristics was first established for each basin separately (the characterizations are separate). The waste characterization basis was established in 2004, using data collected in 2004 and modeling. Therefore, 2004 was established as the baseline period for comparison. As

long as the data from succeeding years did not show a trend that was adverse to the radioactive characterization, existing waste characterization methods could continue to be used to characterize waste for management.

After the basin baselines were established, radiological survey data were collected from each basin and evaluated quarterly. Evaluations of the basin data detected trends but none were adversely affecting the radioactive characterization of the waste. Therefore, the waste characterization remained valid for use. The KE Basin (where material has been removed) showed stability, that is, no trends or shifts. The KW Basin (where radiological material has been received) showed data shifts or trends, but none were outside the acceptable baseline established in 2004.

The use of statistical process control methods to identify trends has been successful in identifying data shifts or trends, and determining if the shift is significant. The trends identified generally align with events occurring in each basin. The determination of significant and adverse trends has eliminated false alarms that would have caused unnecessary corrective actions. The application of statistical process control to monitor radioactive characteristics has been shown to be a robust method that can be applied to many situations where the identification of changing conditions is necessary, such as water monitoring, contamination control, and waste characteristics monitoring.

BACKGROUND

In 1999, two facilities for storing spent nuclear fuel (SNF) underwater at the Hanford Site in southeastern Washington state were authorized under the *Comprehensive Environmental Response, Compensation, and Liability Act of 1980* (CERCLA) to be removed from service, decommissioned, and the facilities dismantled and removed [1]. The KE and KW reactors and their associated fuel storage basins were constructed in the early 1950s and are located in the Hanford 100 K Area near the Columbia River. The fuel basins are large, open-topped concrete pools, which contain demineralized water that is contaminated with dissolved radioactivity and varied concentrations of suspended solids, depending upon the underwater activities being performed. Figure 1 shows a plan view of the KW Basin. The KE Basin is a similar physical configuration. The basins were originally used to store SNF produced by the KE and KW Reactors until the early 1970s, when these reactors were removed from service and the fuel removed from the basins. There after these Basins were recommissioned to store SNF from Hanford’s N Reactor.

In 2000, spent fuel removal from the KW Basin was initiated. In 2002, spent fuel was removed from the KE Basin, and relocated in the KW Basin. All SNF was repackaged and subsequently removed to a dry storage facility located on the Hanford site. By 2004, all the fuel had been removed from both basins.

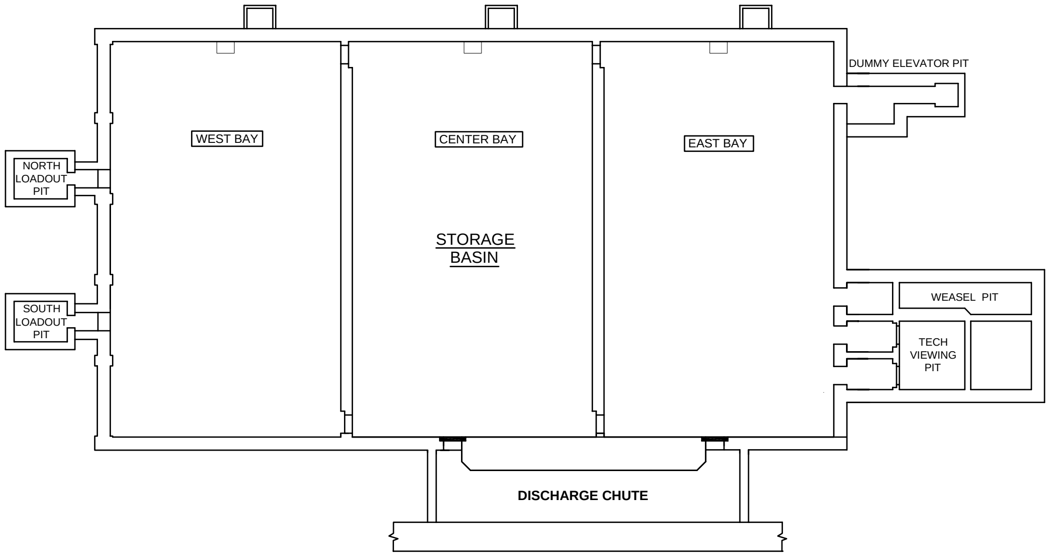


Figure 1. KW Basin plan view

The process of collecting sludge for removal started in 2003 by vacuuming it into vessels submerged in the basins. However, to complete vacuuming in the KE Basin, fuel racks, equipment, and other debris had to be removed to access sludge. Eventually, all the sludge from the KE and KW Basins will be collected in containers submerged in the KW Basin and will eventually be treated and disposed. After all the sludge has been completely removed from the basins, the water will be drained and the superstructures torn down. Lastly, the basins will be stabilized as needed, cut up and/or broken into rubble, and then transported for disposal at a location on the Hanford site.

Equipment, debris, and structures continue to be removed from the basins. These materials, which are radioactive waste, are packaged, treated as necessary, and then disposed in a land-disposal unit located on the Hanford site.

When fuel-removal activities began in 2000, a plan was developed that provided methods and requirements to characterize, treat, and dispose of the solid radioactive waste except for sludge [2]. The characterization, management, and disposal of water, fuel, and sludge is based on sample analysis and is excluded from further discussion in this paper. The waste, consisting of debris, equipment and building structures, must be characterized for radioactivity to properly manage, ship, treat (if necessary), and dispose of the waste. The different classes of radioactive waste have different management requirements:

- Radioactive waste, if determined to be transuranic, must be disposed at the national repository in Carlsbad, New Mexico.
- If the waste is Nuclear Regulatory Commission (NRC) greater-than-class C (GTTC), then it must be disposed at the Hanford low-level waste burial grounds.
- If the waste is low-level NRC class A, B, or C waste, then it is disposed at a CERCLA disposal facility located on the Hanford site.

Because each of these disposal facilities has different requirements, it is necessary to properly radiologically characterize and classify the waste.

The radioactive waste from the basins is characterized using a three step process: 1) measure the gamma dose-rates on the exterior of a package filled with waste, 2) use a dose-rate-to-Cs-137 relationship to calculate the Cs-137 inventory contained in a package, and 3) calculate the inventory of the other radionuclides in the package by multiplying the Cs-137 inventory by the ratio of each radionuclide to Cs-137 established by a distribution for that waste (e.g., above-water debris from KE Basin). Before these characterization steps can be performed a radionuclide distribution (relative concentrations of nuclides) for the waste being generated and dose-rate-to-Cs-137 relationships must be established. The source of radioactive contamination in the basin facilities is the spent fuel that had been stored there. Spent-fuel radioactivity consists of gamma and beta emitters, primarily Cesium-137 (Cs-137) and Strontium-90 (Sr-90); and alpha emitters, primarily isotopes of plutonium and Americium-241 (Am-241). Radioactive distributions for each basin were established based on sample analysis results, gamma assays of some waste containers, and fuel radionuclide inventory data [3]. Each basin exhibited a unique distribution for material removed from under the water and removed from above the water. The relationships of gamma dose-rate-to-Cs-137 inventory were established for various waste packages using modeling.

As the work progresses, it is expected that the radiological characteristics of each basin – that is the distribution – may change as radioactive materials are being moved within and between the basins. It is imperative that these changing characteristics be monitored for changes that might affect the management of the waste. Effective monitoring of radioactive characteristics of the basins will result in properly classifying the radioactive waste, and managing the debris as it is removed.

PREVIOUS MONITORING METHOD

The characterization plan established a method to monitor the radioactive characteristics of the basin facilities by analyzing samples of basin water [2]. Analysis results of basin water were chosen as the performance indicator because basin water is the media delivering contamination to both underwater and above-water material in each basin. Water samples were tested each month for key radioactive constituents including Cs-137 and Sr-90. The ratio of Cs-137-to-Sr-90 concentrations was selected as the indicator to be monitored. Shifts in radioactive characteristics were to be identified by showing that the new data was statistically different from the concentration ratio of Cs-137-to-Sr-90 between 1999 and 2000. A single fixed average and standard deviation values for each basin of the ratio of Cs-137-to-Sr-90 were established using 1999 and 2000 water data. An alarm value of two standard deviations from the average was set for each basin. If the water sample results after 2000 deviated by more than two standard deviations from the 1999 and 2000 average, then corrective action was required before waste could continue to be treated and disposed using the established waste characterization.

In 2003 and 2004, however, the radioactivity concentration ratios of Cs-137 and Sr-90 in the basin water varied tremendously, depending on activities being conducted at the basin. Some basin activities stirred up the sludge and caused the ratio of Cs-137 to Sr-90 in basin water to change. Likewise, when activities ceased, the concentrations of these nuclides would decrease, and the ratio changed again. As a result the Cs-137-to-Sr-90 ratio in the basin water constantly fluctuated outside of the statistical tolerances stated in the previous paragraph, which required that some corrective action be taken. Further, other measures of basin radiological conditions did not show these radical shifts. Therefore, it was concluded that using the Cs-137-to-Sr-90 ratio in the basin waster was not a good performance indicator and basis for waste-management decisions because it was too sensitive to basin activities and did not represent the conditions of the debris being removed. Therefore, a new method to monitor the basins for changes in radioactive characteristics was necessary.

STATISTICAL PROCESS CONTROL METHOD

A different method to monitor the radioactive characteristics of basin debris was clearly needed. This method had to meet specific criteria: accurately identify changes that would affect the management of radioactive waste, provide a stable signal to be monitored, and minimize false alarms that lead to unnecessary corrective action. The statistical process control (SPC) method was chosen because it has been widely used in industry for decades with a proven record of performance. It was originally developed by Dr. Shewhart [4] in the 1930s and further developed by Dr. Deming [5] [6] from World War II through the 1980s. SPC has several advantages:

- It is one of the simplest statistical analysis that separates signal from noise
- The actual data are plotted in a visual manner
- Analysis results are replaceable by different analysts
- False alarms and unnecessary reactions to the latest datum value are minimized
- Trends are identified in a timely manner
- SPC rules detect both small long-term shifts, and large short-term shifts on one chart
- When there is a trend detected, it is likely there will be as special cause found that will be useful
- When there is no trend, it is not likely that a special cause will not be found
- Process tampering and wasted action is minimized
- Can be performed without sophisticated or expensive computer software
- Provides black-and-white feedback on the impact of improvement actions taken.

The application of SPC to a process is achieved via the following steps:

- Identify the business objective
- Define the performance indicators
- Collect performance indicator data.

The business objective has been discussed previously, but is stated here for clarity: monitor the radiological characteristics of the KE and KW Basin facilities to determine when changes that adversely affect the radiological characterization and management of the debris waste removed from the basins exist so that corrective action may be taken. The next two steps implementing SPC are discussed under the headings of the same name.

DEFINE THE PERFORMANCE INDICATOR

A performance indicator must be selected that will identify adverse changes in the characteristics of the radioactive debris waste. An adverse change is one that could result in the waste's being incorrectly determined to be of a waste class that would result in it being managed under less rigorous requirements than required. The radioactive waste classes that are pertinent to KE and KW debris waste in order of higher to lesser management rigor are the following: TRU; low-level NRC GTCC; or low-level NRC A, B, or C. The waste-treatment and disposal regime tolerates errors when a waste is classified to a waste class that has more rigorous requirements than the class it should managed as (e.g., TRU when it is really NRC GTCC). These errors may increase the cost to manage a waste, but do not represent a violation of laws and regulations. Therefore, these errors are not considered adverse changes that would require corrective action.

The KE and KW Basin radioactive relationship that is important for determining radioactive waste class is the ratio of alpha-emitting actinides to Cs-137. Therefore, the relationship selected to be monitored is the ratio of alpha emitters, dominated by the actinides, to gamma emitters, dominated by Cs-137. An adverse change, then, would be the ratio of alpha emitters to beta/gamma emitters increasing significantly. A change in this ratio would be an indication that the radionuclide distribution that was established with previous information may no longer accurately represent the actual ratio to Cs-137. An increase in the ratio would indicate that the distribution to calculate the actinide inventory may under represent the inventory of actinides. A change where the ratio decreases would be an indication that the radionuclide distribution that was established with previous information would result in the calculation of an actinide inventory that may over represent the actual inventory of actinides. The over representation of actinide inventory is an error that the project can tolerate and so is not considered an adverse change that would require corrective action.

The alpha and beta/gamma content in waste can be measured with hand-held field instruments used for radioactive contamination control. The basins are radioactive areas, and so are routinely monitored for fixed and smearable contamination in above-water areas of each facility. The measurement of radioactive contamination on waste material from below the basin water was discarded as a source of data because measurements can only be collected when material is removed from under the basin water which is an infrequent activity.

The project adopted monthly above-water contamination survey measurements of smearable alpha and beta/gamma as the performance indicator data source. More frequent measurements are not necessary as the project needs to identify long-term trends or data shifts. The performance indicator was selected to be the ratio of alpha to beta/gamma results of these surveys. Fixed dose rate measurements were discarded as the performance indicator as it could be affected by background radioactive fields and the smearable measurements were judged more likely to respond more quickly to changing conditions. The performance indicator data will be evaluated at least once each three months [7].

COLLECT PERFORMANCE INDICATOR DATA

Establishing a Performance Indicator Baseline

The KE and KW Basins debris waste characterization basis was established in 2004, using data collected in 2004, historical data, and modeling. Therefore, calendar year 2004 was chosen as the baseline time period for comparison to subsequent years of data.

The monthly reports of radiological surveys during 2004 were examined and smearable contamination results that were usable were tabulated for each facility. Usable results are those that had detectible results for both alpha and beta/gamma measurements of contamination at a sample location. If one or both of the results were less than detection limits then the ratio at that location would be indeterminate and useless for determining a ratio. In some cases, weekly contamination survey reports had to be consulted, as no usable measurements were collected in some monthly survey reports. The ratio of alpha to beta/gamma measurements versus the month when the measurements were conducted at each facility were tabulated for evaluation. At least 30 data points were necessary to provide an adequate statistical sampling of the population to make inferences. The number of KE Basin data points in the baseline year is 56; the number of KW Basin data points, 30.

A chart for each facility was developed by plotting the alpha to beta/gamma ratio against time. The resulting chart shows graphically how the basin facility's radioactive characteristics change over time. In many months, more than one data point exists that was usable so each point is plotted separately for that month. The order in which data of a month are plotted is simply the order in which the data is recorded on the survey reports. The data is then evaluated to identify trends, general data shifts up or down, using rules established in the "*Hanford Trending Primer*" at http://www.hanford.gov/safety/vpp/trend.htm. The data evaluation rules are stated as follows:

- Individual points above the Upper Control Limit (UCL)
- Individual points below the Low Control Limit (LCL)
- Seven points in a row above the average or all below average
- Seven points in a row increasing
- Seven points in a row decreasing
- Ten out of eleven points in a row all above average or all below average
- Cycles or other non-random patterns in the data

- Two out of three in a row that are two standard deviations above the average or two out of three in a row that are two standard deviations below the average
- Four out of five points in a row are outside of two standard deviations above the average, or four out of five points in a row are outside of two standard deviations below the average.

A control chart is constructed by adding the mean, UCL, and LCL to each data plot. The UCL is defined as the mean plus three standard deviations. The LCL is defined as the mean minus three standard deviations. To establish an initial mean, UCL, and LCL, a region of data is visually identified as being stable, that is not showing an obvious trend up or down. For each basin, the initial region started at the earliest time (at the left of the chart) of the data until it is visually observed to clearly shift up or down. For example, the KW Basin data bounced up and down, but in no discernable pattern until the second data point in July 2004, after which the data was consistently lower than the previous data for the next eight data points. The mean, UCL, and LCL were calculated for the initial region encompassing January through the first data point of July 2004. Then, the rules described in the previous paragraph were used to identify clear and maintained shifts or trends in the data from the region immediately prior to it in time.

The KE Basin control chart is shown in Figure 2 which presents both the baseline data and subsequent data collections until September 2006. The baseline data do not show sustained trends or data changes, so all the 2004 data, except two outliers, were used to calculate the mean, UCL, and LCL values of 0.035, 0.124, and 0.006, respectively. Notice that two data points in April, one in July (also 2 out 3 in a row above 2 standard deviations), and one in December were above the UCL, which is an indication of a trend. However, the trends were not sustained, so the data was not treated as separate regions and are not included in the calculation of the mean, UCL, and LCL in order not to skew the data upward. These points are outliers that do not influence the data analysis. The decision rule for an adverse change in the radioactive characteristics of the KE Basin is when a sustained (three consecutive months) data shift or trend that would result in a mean value greater than the baseline mean of 0.035 exists.

The KW Basin control chart is shown in Figure 3, which presents both the baseline data and subsequent data collections until September 2006. In contrast to the KE Basin baseline, the KW Basin data does show sustained data trends or shifts, the first trend being the last data point of July 2004. Regions where sustained data trends exist are charted separately from surrounding regions with the mean, UCL, and LCL being calculated using data of that region only. Three regions of stability were identified in the baseline year, January through the first data point in July, the last data point in July through the second data point in October, and the third data point in October through the end of 2004. The three regions exhibit an alpha to beta/gamma ratio mean of 0.065, 0.016, and 0.032. One data point in October exceeded the UCL which would indicate an upward data trend but as the trend was not sustained the data was not treated as a separate region and is not included in the calculation of the mean, UCL, and LCL in order not to skew the data upward. This point is an outlier that does not influence the data analysis. The decision rule for an adverse change in the radioactive characteristics of the KW

Basin is when a sustained (three consecutive months) data shift or trend would result in a mean value greater than the maximum baseline mean of 0.065 exists.

Evaluation of Performance Indicator Data

The results of smear measurements are tabulated in three-month increments and added to the control chart for each basin as described for preparing the baseline control charts. Again, the rules to identify trends are applied, if regions of sustained trends are identified, then the mean, UCL, and LCL are recalculated for that region and plotted on the control chart.

The KE Basin monitoring results of 2005 through September 2006 were plotted on Figure 2 with the baseline year 2004. The data collected and plotted after the baseline year did not show any sustained trends or data shifts though the data fluctuated around the baseline mean. So a data trend was not identified, adverse or otherwise, for the KE Basin radioactive characteristics.

The KW Basin monitoring results of 2005 through September of 2006 were plotted on Figure 3 with the baseline year 2004. After the baseline year, five more regions were identified as exhibiting data trends or shifts, with the mean ranging from 0.014 to 0.064. On four separate occasions, one or two points exceeded the UCL, which indicates a trend; however, as these trends were not sustained, the data is not treated as a separate region and are not included in the calculation of the mean, UCL, and LCL so that these parameters are not skewed upward. These points are outliers that do not influence the data analysis. So far then, data trends have been identified, but no adverse trend has been identified for the KW Basin as the calculated maximum mean of post 2004 data is 0.064, which is less than or equal to the maximum mean in the baseline year of 0.065.

The performance indicator for the KE Basin facility has remained stable probably because during the baseline and operating periods no new sources of radioactive material had been added. The stability of the performance indicator infers that the radioactive characteristics are also stable. Radioactive materials such as fuel and above-water and underwater debris have been removed from the KE Basin and sludge that already existed in the basin was collected and relocated inside the basin.

Figure 2. K East Basin Control Chart- 2004 through 2006

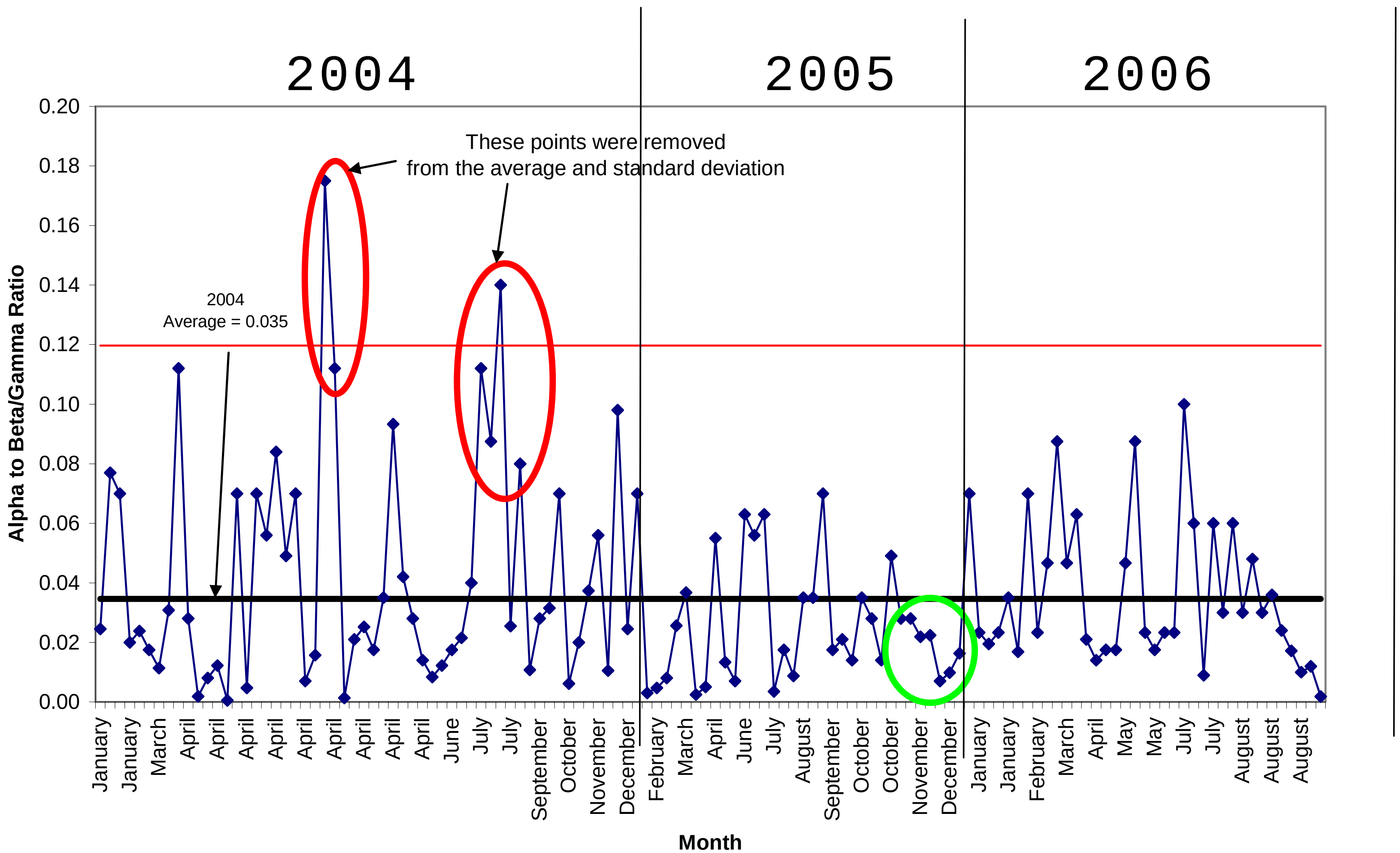
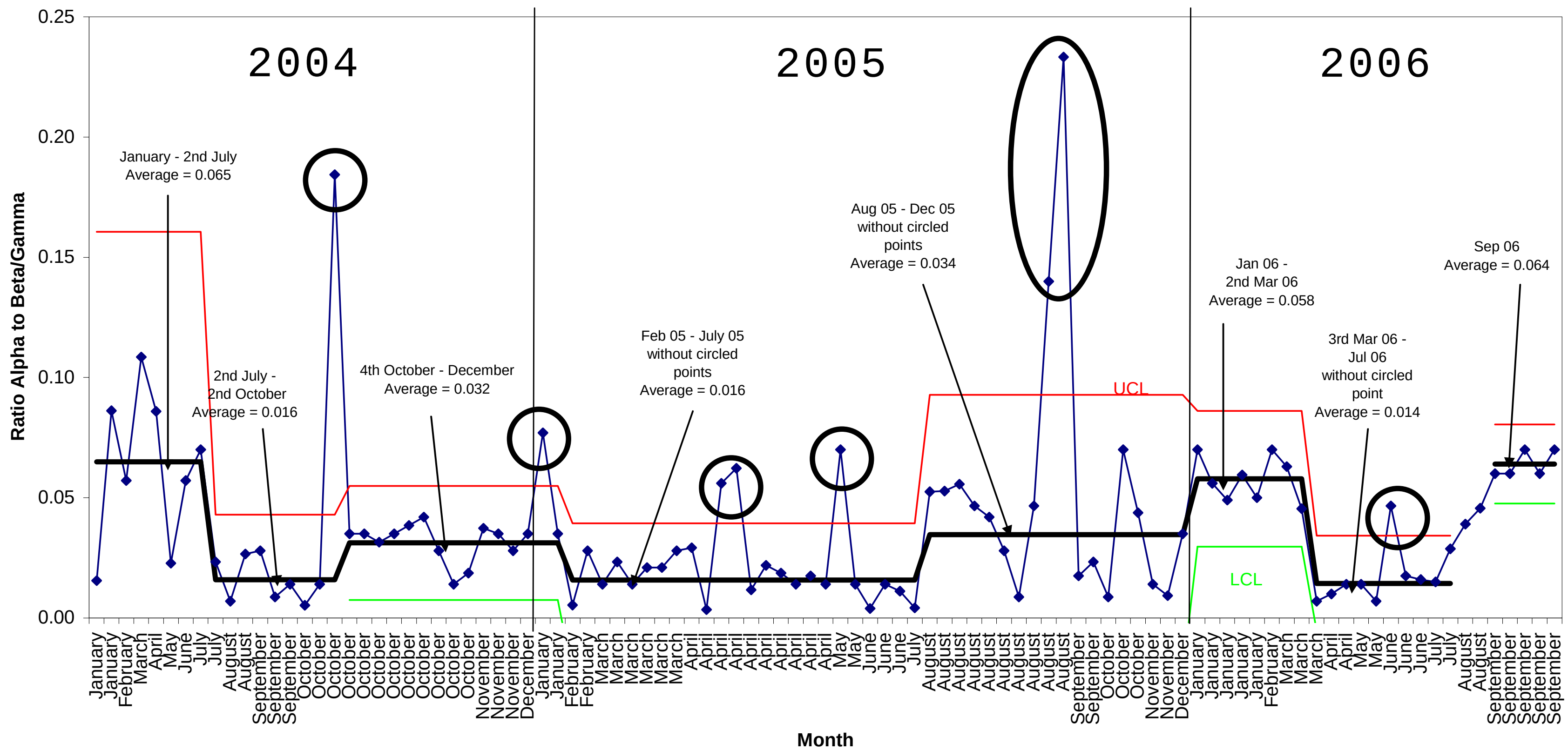


Figure 3. K West Basin Control Chart- 2004 through 2006



In contrast, the KW Basin exhibited multiple data trends. Like the KE Basin, the activities in the KW Basin included removing SNF and above-water and underwater debris. Unlike the KE Basin, however, the KW Basin also received and packaged SNF during the monitoring period and experienced an increase in sludge inventory. Before the KW Basin received and packaged SNF, it contained very small amounts of sludge. The SNF packaging and sludge-movement activities in the KW Basin align with the higher mean alpha to beta/gamma ratio measurements. Fuel processing and scrap processing occurred from January to July 2004 and October to December 2004, which corresponds to regions of high mean alpha to beta/gamma ratios. Sludge-movement activities such as pumping and vacuuming occurred from August 2005 to February 2006 and July to September 2006, which again corresponds to regions of high mean alpha to beta/gamma ratios. The control chart identified these activities that create increased contamination of the basin facility using the rules that were previously stated. Sludge movement and fuel and scrap fuel processing represent conditions of higher alpha contamination due to the release of radioactivity under the basin water. For example, by applying the rules to the KW Basin August data, it was clear that the performance indicator had shifted up, as multiple points were above the UCL of 0.04 established for the region of data February to July 2005. Therefore, a data shift or trend created a new region, over the period of August to December 2005, the mean, UCL, and LCL were calculated and plotted on the control chart.

The performance indicator in the KW Basin tracked with activities that would release radioactivity into basin water. However, the performance indicator in the KE Basin did not track with activities. The KE Basin did not track because the large sludge inventory continually releases radioactivity into the basin water throughout the monitoring period, which masked these activities. As there have been no adverse trends of the performance indicator (i.e., increasing alpha to beta/gamma ratio above the baseline values), the radioactive distribution remains representative of the waste and valid for use to calculate the radioactive inventory of containers of radioactive waste.

A process improvement to the monitoring process is under consideration. The improvement is a modification of the data collection process to provide more usable data each month. As discussed previously, survey locations often do not yield usable data as one or both of the measurements at a survey location are less than detectible. The many not detectible measurements are due to repetitive smears being taken from the same spot in a certain location or smears being taken in less active areas within the basins where contamination would not accumulate. More usable data could be obtained if the project used weekly survey reports or a new survey plan was developed that targeted active locations in the basins where detectible measurements are more likely to exist and are located at likely sources of contamination of the basin facilities.

CONCLUSIONS

A SPC method to monitor the radioactive characteristics of the KE and KW Basins has been successfully implemented for almost two years. While evaluations of the basin data detected trends, none were identified as adversely affecting the radioactive characterization of the waste. Therefore, the waste characterization remained valid for use. The KE Basin (where material had been removed) showed stability, that is no trends or shifts. The KW Basin (where radiological material had been removed and new material added) showed data shifts or trends, but all were inside the acceptable baseline established in 2004. A process improvement to modify the way the data is collected is being considered to obtain more usable measurements. The use of statistical process control methods to identify trends has been successful in identifying data shifts or trends, and determining if the shift is significant. The trends identified generally align with fuel packaging and sludge movement activities occurring in the KW Basin. No trends were detected in the KE Basin. The stability of the KE Basin data is probably due to the large sludge inventory that is a source of contamination that masks contamination created by work activities. The KW Basin does not have this large sludge inventory. The SPC process has eliminated false alarms that in the past would have caused unnecessary corrective actions. The application of statistical process control to monitor radioactive characteristics has been shown to be a robust method that can be applied to many situations where the identification of changing conditions is necessary, such as water monitoring, contamination control, and waste characteristics monitoring.

REFERENCES

1. U.S. Environmental Protection Agency, *Declaration of the Record of Decision for DOE Hanford 100 Area, 100-KR-2 Operable Unit,* 1999.

2. J. H. Zimmerman, HNF-6495, *Sampling and Analysis Plan for K Basins Debris*, Revision 1, 2001.

3. J. L. Westcott, HNF-6273, *Data Quality Objectives Process for Designation of K-Basins Debris,* Revision 0, 2000.

4. W. A. Shewhart, Economic Control of Quality of Manufactured Product, 1931.

5. W. Edwards Deming, The New Economics, 1994.

6. W. Edwards Deming, Out of the Crisis, 1986.

7. A. L. Prignano, HNF-6495, *Sampling and Analysis Plan for K Basins Debris,* Revision 2, 2005.