

Operational Changes & Impacts on LLW Scaling Factors



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Operational Changes & Impacts on LLW Scaling Factors

1008017

Final Report, December 2003

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This report was prepared by

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This report describes research sponsored by EPRI.

The report is a corporate document that should be cited in the literature in the following manner:

Operational Changes & Impacts on LLW Scaling Factors, EPRI, Palo Alto, CA: 2003. 1008017.

REPORT SUMMARY

In accordance with NRC regulatory guidance, utilities may use correlations between long-lived, difficult to measure radionuclides and more readily measured radionuclides for characterization of low level wastes. Derivation of scaling factors has been an ongoing process at nuclear power plants for 20 years. During that time, each plant has accumulated, at considerable expense, a substantial body of radiochemistry analyses. Many plants are looking for opportunities to reduce these expenses and take advantage of the experience already gained. This study examines the impact of operational changes on scaling factors to determine their extent, using actual data collected from operating plants

Background

Since 1984, the industry has introduced several significant changes in plant water chemistry management, including zinc injection to limit the incorporation of Co-60 in the corrosion layer, and hydrogen water chemistry to displace oxygen and limit stress corrosion in BWRs. In PWRs, the changes include varying lithium concentrations to control formation of corrosion layers. More recently, a number of PWRs experimented with zinc injection into primary coolant to limit the potential for stress corrosion. In addition, both BWRs and PWRs have experienced fuel failure episodes and extended outages, and have undergone major decontaminations. It is generally believed that such changes and transient conditions cause temporary or permanent changes in scaling factors, but to date no attempt has been made to show which scaling factors were affected or the extent of the effect. Similarly, utilities have observed impacts on scaling factors, particularly because of changes in fuel design. This report discusses the impact of these operational changes on scaling factors.

Objectives

- To determine, within the precision constraints defined by the NRC and the sensitivity of the overall process, if operational changes cause impacts which are observable and quantifiable using standard statistical methods.
- To provide guidance incorporating consideration of these changes for planning and updating radiochemistry sampling and scaling factor sampling.
- To examine the issue of declaring certain ratios constant and to present draft criteria for such declarations.

Approach

The project team collected data from several candidate nuclear power plants. Ideal candidates were those with a continuous sampling record and identifiable events that the team could reasonably tie to a period of operation. The data were entered into a database and evaluated using 10CFR61 Sample Analysis Program. The project team modified the program to allow data

groupings covering periods before and after the introduction of the change. They compared these groupings to identify statistically significant differences and determine the extent of the difference. The report presents evaluations for critical scaling factor ratios for eight plants including four BWRs and four PWRs.

Results

The results of this investigation show that operational changes do have the capability to temporarily and permanently impact scaling factor values. In many cases, other ongoing changes can obscure these impacts. However, the impacts are generally within the sensitivity of the overall process, as well as within the precision constraint defined in NRC guidance. None of the specific changes examined would impact scaling factors to the extent that supplemental sampling would be required.

Constant scaling factors for activation products and transuranic radionuclides are a viable option when supported by the database. Utilities should base such determinations on the longest supportable sample record leading up to the current values. They should also consider trends to determine the necessity and timing for continued verification. Plant managers should also define bounding values for the scaling factor for comparison with future confirmatory analysis.

EPRI Perspective

EPRI has focused long-term effort to supplying the technical bases for the use of LLW scaling factors for its nuclear power members. The result has been significant cost savings to the industry obtained through the reduction of unnecessary sample analysis. This project determined that for plants using an accepted LLW Scaling Factors program, utilities should not need to require additional sampling because of typical operational changes.

Keywords

Radioactive waste LLW Correlations Scaling factors, constant Characterization

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1 INTRODUCTION

Scaling factors are defined as correlations between difficult to measure (*dtm*) radionuclides and corresponding radionuclides (*key*) that can be readily measured by gamma spectrometry. The correlations are developed from radiochemical analysis of samples collected from radioactive waste streams. The radiochemical analyses are performed by an offsite laboratory since most generators lack the specialized equipment needed to conduct the measurements. The scaling factor is then applied to new material to estimate *dtm* nuclide activities from local measurements of *key* values.

Scaling factors have always been used in some form for estimating the quantities of difficult to measure radionuclides in radioactive materials. The use of scaling factors became more formalized in 1983 with the introduction of the Low Level Waste Policy Act and the accompanying NRC regulations in 10CFR61. Utilities in the United States are nearing 20 years of experience in collecting sample data for long lived radionuclides and are beginning to look for ways to take advantage of this experience to reduce future sampling expenses. Experience can be used to advantage by trending data collected over a period of many years to develop statistically valid estimates that cover a range of operating conditions. Data collected at many plants demonstrate a pattern or signature distribution of radionuclides. By a rigorous examination the data, waste streams demonstrating similar ratios can be combined to reduce sample frequencies, ratios shown to be constant between wastes streams may be excluded from general analysis or examined in one or two samples per cycle. Sample cycles can be extended by showing that the range of operating conditions encountered have had little effect on the scaling factors determined from the samples collected.

Regulatory Basis/ Rules

Publication of 10CFR61 was followed directly with the publication of the NRC Branch Technical Position (BTP) on Waste Classification. The BTP placed the burden on licensees (e.g. NPP Operators) to "carry out a compliance program to assure proper classification of the waste." The BTP states further that acceptability of the compliance program would depend on NRC's determination that the licensee had made a reasonable effort to obtain a realistic representation of the distribution of radionuclides in the waste and that a reasonable target for determining measured or inferred concentrations would be estimated within a factor of 10.

NRC outlined four methods that could be used for monitoring and determining radioactivity content in various waste forms. These included:

Materials Accountability - Applicable to cases where licenses receive and possess a limited number of radioisotopes in know concentrations and activities.

Introduction

Classification by Source - Applicable to cases were licensee has knowledge and control of the source of the waste.

Gross Radioactivity Measurements - Applicable to all types of waste given a correlation on a consistent basis with a distribution of radionuclides within and particular waste stream.

Direct Measurement of Individual Radionuclides - Applicable to all waste types and allows the use of an inferential measurement program to control costs and exposures resulting from sampling.

The method found to be most suitable for wastes generated at nuclear power plants is that of direct measurement of specific radionuclides. Gross radioactivity measurements (dose rates) are also used to characterize waste materials where the spectrum is known but where it is not possible to obtain a representative absolute concentration by measurement. NRC specifies that for the gross radioactivity measurement method that the spectrum be initially determined by direct sampling and radiochemical analysis and periodically verified. To reduce radiation exposures incurred by sampling and costs of implementation, the NRC allowed the use of correlation (scaling) factors to generalize the relations between radionuclides that required radiochemical determination and those that could be measured using equipment normally available to the generator.

Both the direct measurement method and the gross radioactivity measurement method rely on spectra determined by radiochemical analysis of samples and may or may not involve the use of scaled values to complete the spectrum.

NRC recognized in the Branch Technical Position that concentrations would likely be determined based on sampling of waste streams. NRC also stated that <u>correlation factors should</u> <u>be determined on a facility and waste stream specific basis</u>. This meant that each plant had to conduct its own sampling program. Sample data from similar plants or generic data was not sufficient to comply with NRC's requirements. This is still true and does not show any signs that it will be changed in the near future.

The over-riding criteria put forward by the NRC is that of *reasonable assurance* that a system of classification using correlations [or any other method] will result in accuracy of estimates within a factor of 10. This criterion was initially stated in the 1983 BTP and has been reinforced in just about every discussion subsequent to that. The demonstration of the method used is left primarily to the generator/licensee.

The NRC has also maintained that the single sample can be used to define scaling factors if it is considered to be <u>representative</u> of the overall waste stream. The concept of a representative sample is very important to understanding the NRC position. The determination of what is representative is left up to the generator's authority. It may be representative by default because it was drawn from the specific waste stream where it will be applied. It may be representative because the scaling factors derived from it compare within a factor of 10 of those derived from a sample taken during the previous operating cycle or with some composite values derived from previous sample cycles. Whether a sample is representative also depends on the sampling process itself. For most waste streams, small samples are drawn from large volumes of material. Activities vary over the length of a filter and significant changes in spectrum can be observed if a smear sample is collected in place of a coring or a clipping. Similarly, for resins, since they are

often mixed in disposal containers, the observed spectrum will depend on the degree of mixing, at what stage of processing collection occurs, and the size of the sample.

The BTP directed that confirmatory analysis of scaling factors (or correlations) should be performed on at least and annual basis for most Class B and C wastes, and at least a biannual basis for Class A wastes. Confirmatory analysis includes collection and radiochemical analysis of new samples. Use of scaling factors may be extended if the new samples indicate that continued use of the previous scaling factors would still provide reasonable prediction of actual concentrations within a factor of 10. In addition, the NRC indicated that re-analysis of *dtm* radionuclides should be performed when¹ "there is reason to believe that facility or process changes may have significantly altered (e.g. by a factor of 10) previously determined correlations of gross radioactivity measurements".

Thus, the NRC established a "precision" standard for radiological characterization and the use of scaling factor for that purpose. The determination should be neither to low or excessively conservative. It acknowledges differences between plants and differences between streams within each plant. While the process is not exact, it is expected that when it is applied globally that on average it will reflect the actual disposal source term. If the process were made more specific implementation would be neither economical nor consistent with ALARA objectives. If it were made more global, it would risk missing outlying generators that could by themselves effect the overall source term.

Additional Guidance on the Implementation of 10CFR61

To facilitate the implementation of 10CFR61 and to respond to specific questions, NRC published "Additional Guidance on the Implementation of 10CFR61" in December of 1983. Among the topics, NRC stated that written procedures and controls must be in place for classifying waste. However, the NRC also indicated that related activities need <u>not</u> be included in corporate level quality assurance programs for nuclear safety related items, rather classification activities should be consistent with the recommendations for radwaste treatment systems in Regulatory Guide 1.143. The quality assurance issue is also addressed in the more recently published 10CFR20 Appendix G.

Another issue addressed in the "Additional Guidance" document was related to the difficulties in scaling H-3 and C-14. The following is quoted from questions and answers distributed with the Additional Guidance.

"Tritium and C-14 are nuclides that are difficult to scale. An acceptable approach to determining waste concentrations for these nuclides would be to assume that the waste contains the same concentration that was measured in the sample assay.

In most cases Tc-99 and I-129 will be below the lower limit of detection (LLD) in routine radwaste samples. In these cases, it is acceptable use the LLD as the assumed waste concentration. It is also acceptable to develop a correlation factor based on the

¹ BTP on Radioactive Waste Classification, May 1983

LLD for the analysis. A fission product should be used as the base nuclide for these correlations."

In March 1986, NRC issued , IE^2 Information Notice 86-20 which reinforced its position against using generic and non-facility specific data as basis for scaling factor determination. The complaint from the NRC was that other methods could not provide reasonable assurance that inferred radionuclide concentrations would be accurate within a factor 10. Concern was expressed that low estimates would be non-conservative and systematic gross estimates could artificially limit the use of the disposal facility. Attachment 1 to the notice included a list of which waste streams warranted the determination of unique scaling factors. These are listed in Table 2-1.

Table 1-1 Waste Streams Identified in IE Notice 86-20

Pressurized Water Reactor	Boiling Water Reactor
Primary Purification Filters	Cleanup filters/Resins
Primary Purification Resins	Condensate Polishing Resins
CVCS Evaporator Bottoms	Radwaste Ion Exchange Resins
Radwaste Polishing Resins	Dry Active Wastes
Secondary System Wastes (filters and Resins)	
Dry Active Wastes	

It was also noted in Attachment 1, that as a sample analysis history of facility waste streams is compiled, licensees may choose to determine new scaling factors based on the most recent sample analysis results or may combine the latest analysis with those previously obtained to refine the scaling factors currently in use providing that the old results remain representative of the stream. (*This clarification provides some sanction for averaging data between sample cycles given stable operating conditions or at least conditions which do not affect the ratios being evaluated.*)

The IE notice took particular issue with those plants that attempted to use a single set of scaling factors defined by selecting the most conservative ratio from each stream sample and combining these into a set of scaling factors that would envelop all of the streams. The concern stated by the NRC was that in order to meet 10CFR61 disposal objectives it might be necessary to set inventory restrictions on certain radionuclides. The method of enveloping grossly and systematically overestimated the quantities of radionuclides and could result in under utilization of disposal capacity and higher disposal costs.

² IE Notices are issued by the NRC's Inspection and Enforcement Branch. These are usually issued to highlight some problem that is observed by inspectors to require regulatory clarification.

Previous work by EPRI /Data Collections, Guidance Studies

Radionuclide Correlations in Low-Level Radwaste, Electric Power Research Institute, Palo Alto, CA, EPRI - 4037, June 1985

This study examined the viability of indirect determination of difficult-to-measure radionuclide concentrations the more easily measurable radionuclides. Special sampling and scaling factor development were performed at 2 BWR and 2 PWR plants as demonstration of the methodology. This report introduced the geometric mean and dispersion as the primary evaluation tool for scaling factor analysis, including a series of "probit"³ plots to highlight the log-normal distribution. In addition data was collected in a survey of operating plants and generalized scaling factors were derived for principal waste streams.

Updated Scaling Factors in Low Level Radwaste, Electric Power Research Institute, EPRI-5077, March 1987.

This study provided an update to the work performed for NP-4037. It included an expanded database of 1300 samples. Using results from the data collected, the study proposed "generic" scaling factors for Fe-55, Ni-63 and TRU radionuclides relative to Pu-239 based on plant type.

Radsource Computer Program

The RADSOURCE computer program consisted of an updated version of the Vance & Associates ISOSCALE computer program. RADSOURCE is based on trending of reactor coolant radio-iodine to distinguish the source of fission products in reactor coolant between tramp fuel and actual defects in the fuel. This information is used to predict correlations between various fission products including Sr-90, Tc-99, and I-129. These correlations are offered to replace radiochemistry measurements. The update from ISOSCALE to RADSOURCE replaced the system of decontamination factors between streams with a more flexible system of transport factors based on waste stream specific radiochemistry reports. To support the development of the transport and removal factors (TRAFs), new data was collected that expanded the EPRI data base to more than 3000 samples. A determined effort was made to develop TRAFs between primary (or RWCU) resins and other waste streams. The major difficulty encountered was that there was no reliable data for comparison for either Tc-99 or I-129.

In conjunction with the RADSOURCE development a separate report was prepared presenting the results of the TRAF evaluations, updated scaling factors, and a number of observations relating to expected scaling factor response to plant conditions. Also addressed in this report were general impacts on characterization reliability including dose to activity analysis, sampling methods, laboratory handling of samples, and representative sampling.

Low Level Radioactive Waste Characterization Guidelines (EPRI TR-107201; 1996)

³ A plot of the log ratio versus the percentage of values lower than the current value. A linear plot indicates normal conditions.

Introduction

This report aimed to consolidate the experience gained by EPRI and the industry. Using data and information included in previous reports and a reanalysis of the EPRI database, guidelines and recommendations were presented for defining waste streams, conducting a sampling program, addressing laboratory issues, evaluation of scaling factors, methods for classification, and what to report in disposal manifests. The report was prepared by a committee formed of characterization specialists from more than 20 power plants working with consultants and editors.

Utility Use of Constant Scaling Factors (EPRI TR-109448; 1998)

This report identified several constant scaling factors utilities can use to reduce their sampling and analysis requirements for compliance with 10CFR61 waste classification regulations. Plant wide and waste stream specific constant scaling factors were identified in the study. Utilities can easily establish procedures for verifying and using these factors to decrease sampling and analysis costs, as well as to reduce personnel exposure.

Constant Scaling Factor Objectives

As noted above, collection of "10CFR61 samples" has been ongoing for the last 20 years. Plants with effective tracking and trending programs are observing a diminishing return on the value of these samples. For activation products and transuranic ratios, new samples tend to reinforce past observations and do not result in significant changes to the scaling factors in use. For I-129 and Tc-99 measurements, no new information is provided since the reported values reflect either the equipment limits of the laboratory or a detection limit specified in the laboratory agreement. For H-3 and C-14, no previous evaluation has ever developed a generalized correlation with Co-60 or Cs-137. An option, in this case, is to use the concentration from the laboratory as a constant concentration and avoid scaling to a key gamma. This leaves two scaling factors that can be meaningfully impacted by new data. These are Sr-90 and Pu-239⁴ that can be effectively scaled to one or another of the key gammas.

Since 10CFR61 sample collection started, there has been a relatively dramatic improvement in fuel performance in the nuclear power plants. Fuel failures have become increasingly rare. As a result, it has become often impractical to use Cs-137 as a key radionuclide since it is reported at the detection limit and not viable as a key and, in fact, not viable in a second order scaling factor relation. This is because the reciprocal of '<1' is '>1'. A scaling factor with an LLD in the denominator will under predict, perhaps by orders of magnitude, the scaled radionuclide. Along with the improved fuel performance, the related advances in materials used in fuel construction can also have important impacts on scaling factors.

Ce-144 has always been problematic for use as a *key* radionuclide. As a fission product, it is the best predictor for TRU. However, it is most often not reported in waste samples and when it is reported, it is often unreliable. Again, if it is reported as limit of detection, it has no use in

⁴ Pu-241 is a viable alternative as the 'key' TRU since it is present at the highest concentration and is a beta-photon emitter rather than an alpha emitter.

scaling factors. Since it is rarely observed in local gamma spectra, it has almost no utility as a *key*.

So, given that so little can be derived from the radiochemical samples, there is some motivation to focus the process so that it will provide new and valuable information for LLW characterization without generating unnecessary expense. Much of the analysis is useless because the measurement can not be achieved or unnecessary because the correlations are already well known. It should be acceptable to omit or limit the repetition of these procedures. The NRC BTP, however, requires an annual update of the scaling factor correlations. The interpretation here is that a new set of samples is collected and full set of radiochemical analyses are performed. Perhaps it is sufficient to meet the intent of this guidance, that is, that the program for developing correlations is maintained current and consistent with expected concentrations in the LLW being generated and disposed. This objective can be achieved with confidence in the scaling factors used. Confidence can be achieved through projections from a history of measurements and understanding of the effects of plant changes on the correlations.

2 STUDY APPROACH

Project Scope

The intent of this examination is to investigate at the plant level the extent that changing conditions have contributed to changes in plant scaling factors. In order to proceed with the examination, it is important that the plants included in the investigation have an adequate sample record to support the review. It is expected that all plants in the United States have undergone at least one significant change. Each plant included in the review should have available one sample per cycle for each of the four major process streams (i.e high activity resins, filters or filter media, low activity resins, and DAW) dating back four cycles prior to the introduction of the change. This is to ensure sufficient data to calculate a variance.

Steps include

- 1. Survey plants for availability of sample records, and interest in participation.
- 2. Select plants for review.
- 3. From each participating plant, collect sample data and operating history information
- 4. Develop a time line for the change in conditions that can be compared with the sample record.
- 5. Examine the trended scaling factors to see if a change in scaling factors can be tied to the operational change. The analysis will consist of calculating a trended scaling factor value for the period before the change and for the period after the change. Where differences are observed, additional tests will be performed to determine, on the basis of the observed variance, if the difference is significant.

Scaling factors examined will be those that are readily determined from radiochemical data including transuranics, Sr-90, and FE-55. No attempt will be made to reconcile impacts on Tc-99 and I-129 since radiochemical results do not reliably report these concentrations. Fe-55 is chosen as a surrogate for activation products since it is observed to be more sensitive to plant changes and respond more definitively. Ni-63 was observed to be less responsive.

Operational Changes

There are a number of different types of changes in plant conditions that can potentially affect scaling factors. At this point nearly all of the changes have been experienced at one plant or another. Some of the changes considered are listed below:

Study Approach

BWR Zinc Addition

Zinc is added to reactor coolant to reduce Co-60 activity in scale deposits. The zinc displaces cobalt in the scale layer thereby reducing occupation exposures around system piping. The effect, at least initially, is that the cobalt is mobilized in the coolant and removed via the reactor water cleanup system. Assuming that there is a relatively constant production rate of Co-60, and a corresponding exchange rate with the scale layer, it would be expected that there would be no long term impact in the concentrations of Co-60 in the radwaste systems. (It may also be that the Co-60 held in the corrosion layer is only a small fraction of the total Co-60 released, in which case there wouldn't be a noticeable difference.)

PWR Zinc Addition

The primary purpose for zinc addition in Westinghouse PWRs is to reduce the potential for inter granular stress corrosion on the primary water side of Inconel 600 steam generator tubes. Reductions in cobalt activity in the corrosion layer as a result of zinc substitution is a secondary benefit. From the stand point of scaling factor impacts, the secondary process turns out to be of primary importance. Issues related to this zinc addition are basically the same in PWRs as in BWRs.

BWR Hydrogen Water Chemistry

In this case, hydrogen is injected into the primary coolant. The purpose of the injection is to reduce free oxygen in the reactor coolant (and other anions). Free oxygen contributes to stress corrosion cracking. The changes to reactor coolant chemistry are subtle from the perspective of radwaste processing and again are expected to have little impact on scaling factors.

BWR Iron Injection

Iron is injected to maintain a minimum iron concentration in reactor coolant. It was found that when iron concentrations dropped below certain levels buildup of activity in hotspots was increased. The iron displaces other elements that might settle into the hot spots. The problem is more pronounced in plants with deep bed demineralizer systems which are more effective in stripping iron. Injection of iron will assure a minimum concentration of elemental iron that could result in an increase in Fe-55. The extent of the impact would depend on how low the iron was prior to implementation.

PWR Lithium Injection

Since PWRs use a substantial amount of Inconel, there is a major concern with amount of nickel released through corrosion to the reactor coolant. Addition of LiOH, lowers the reactor coolant pH and reduces corrosion rates. The reduced corrosion rate limits the amount of Ni-58 in the coolant which, in turn, lowers the amount of Co-58 available to deposit on primary surfaces. Lithium injection was started prior to the period covered by the data available for this review.

Decontamination Efforts

Decontamination is another process used to reduce occupational exposures. Chemical decontaminations usually focus on primary piping and steam generators in PWRS and on recirculation piping in BWRs. Some full system decontaminations have been performed. The decontamination removes deposited scale and results in a burst of activity collected in processes used to cleanup the decontamination solutions. The decontamination waste is expected to be batch sampled and would not be evaluated using scaling factor methods. The decontamination itself would be expected to impact the amount of contamination available for transport within the system and it should be assumed that there will be a transitory effect on scaling factors relating to radionuclides found in the scale including activation metals and transuranics.

Fuel Defects

Fuel defects are the most generally recognized events impacting scaling factors. This subject has been discussed in previous EPRI reports. The opening of a defect is pretty readily observable through reactor coolant chemistry measurements but may not be immediately seen in scaling factors. Small defects will impact the Cs-137 concentration in reactor coolant. If Cs-137 is used as a key for Sr-90 or TRU, reductions in the scaling factors are expected. If Co-60 is used as the key, no impact is likely. Larger defects will effect the release rates of all fission products and TRU and will impact Co-60 scaling factors, perhaps more strongly than those related to Cs-137.

Changes in Fuel Design

Most of the changes related to fuels currently have taken place prior to in the early stages of scaling factor practice. These changes, including the trend to smaller pin sizes and varying cladding materials have resulted in improved fuel performance and less observable Cesium 137 in LLW streams. Since a least the early 1990s, there has been a steady trend within the power plants to rely more strongly on Co-60 based scaling factors. When a new fuel design or type is introduced, it may take several cycles to fully change over the core. With the premium on fuel storage space, there is enough motivation to endure some the presence of some defects or a reconstitution process in lieu of a full core change out. As a result there is no clear demarcation of the change. Cesium just gradually disappears.

Extended Shutdowns

An extended shutdown can occur as a result of major maintenance, fire, flood, or imposed as a result of regulatory issues. During an extended shutdown, the fuel remains within the plant but may be off-loaded from the reactor. The burden of processing primary water shifts to the fuel pool cooling system. Because the reactor isn't being operated, there is little additional activation occurring and little production of fresh fission products. The expected impact on scaling factors is a relative loss of Co-60 and other shorter lived radionuclides due to decay during the shutdown period. Gradual purging of volatile or otherwise mobile radionuclides is also expected.

3 ANALYSIS METHODS

Statistical Methods

Statistical analysis will focus on methods traditionally used in the evaluation of scaling factors. These include primary the geometric mean and dispersions as measures of the scaling factor and the consistency of the value. Previous studies have examined the applicability of geometric treatments and shown that the data exhibit a log-normal pattern supporting the use of a geometric distribution. Additional tests include pooled variance and a regression trend test. These methods and primary equations are presented below.

Geometric Mean

If the individual ratios are defined by

$$\boldsymbol{\chi}_{i} = \left(\frac{dtm}{key}\right) from \, sample \, i \tag{3-1}$$

The geometric mean ratio is defined by nth root of the product of n values

$$\boldsymbol{\chi}_{g} = \left(\prod_{i=1}^{n} \boldsymbol{\chi}_{i}\right)^{1/n}$$
(3-2)

or in log terms, the antilog of the average log value

$$\chi_g = \log^{-1}\left(\sum_{i=1}^{n} \frac{\log(i_i)}{n}\right)$$
(3-3)

Dispersion

Geometric dispersion (d) is defined as the antilog of the variance

$$d = anti \log \sqrt{S^2}$$
(3-4)

3-1

Analysis Methods

where s is the variance defined by the equation

$$s^{2} = \frac{\left(\sum_{i=1}^{n} \log(x_{i})\right)^{2}}{n-1} - \frac{\left(\sum_{i=1}^{n} (\log(x_{i}))^{2}\right)}{n(n-1)}$$
(3-5)

Since the dispersion acts as a factor on the log values, its impact on the geometric mean is multiplicative. A dispersion of 2 would represent variations a factor of 2 times the geometric mean or $\frac{1}{2}$ of the geometric mean. Normally for scaling factor analysis the prediction standard is that the scaling factor predict true values within a factor of 10. This would be true if the dispersion is limited to 5.

Pooled Variance

Pooled variance provides a mechanism for testing the means and variances of different data sets to determine if the data is from the same population. It is used in the Sample Analysis Program to evaluate the feasibility of combining waste streams for sampling purposes. In this case the means of two data sets are compared against a test statistic to determine if there is a statistical difference in the data. The test statistic is taken from Students t-distribution. Basically, given the number of samples and variance in the data, it is determined if the data are from the same population and there is no statistically significant difference even though the particular geometric means and dispersions may be different. Pooled variance (s_b^2) is calculated from the following equation. (Subscripts refer to the individual data groupings where n is the count and s² is the variance.)

$$S_{b}^{2} = \frac{(n_{1}-1)S_{1}^{2} + (n_{2}-1)S_{2}^{2}}{(n_{1}+n_{2}-2)}$$
(3-6)

The test statistic is defined by

$$test = \frac{\left|\mu_{1} - \mu_{2}\right|}{S_{b}\sqrt{\frac{1}{n_{1}} + \frac{1}{n_{2}}}}$$
(3-7)

Where: μ_1 and μ_2 are the population means for the two data groupings

If the test value is less than the Students t-value for the given degrees of freedom, defined by n1+n2-2, it cannot be concluded that the means are different. The higher the dispersion of either data set the less likely it is that there will be distinguishable difference.

In order to isolate the comparison, data is limited to the period comprising 4 cycles before and four cycles after the introduction of the change. In several cases where new chemical processes have been introduced, there is not sufficient data following the change to obtain a comparison.

An artificial value (R) is introduced into the test statistic to quantify the difference between the means. The test expects the two ratios to be identical. If $\mu_1 = \log (m_1)$, then $m_2/m_1 = 1$ if $\mu_1 = \mu_2$. If the ratio of m_2/m_1 equals 3, the test condition would be met by subtracting log(3) from the absolute difference of the log ratios.

$$test = \frac{\left|\mu_{1} - \mu_{2}\right| - \log(R)}{S_{b}\sqrt{\frac{1}{n_{1}} + \frac{1}{n_{2}}}}$$
(3-8)

To quantify the differences, the value R is solved for. If R equals 1, the means are statistically the same. If R is greater than 1, it represents the factor by which the means are different. For a two tailed test (2-s), an R greater than 5 would necessitate re-evaluating the scaling factor to assure a factor of 10 precision. For the purpose of this examination any R value greater than 1 will be interpreted as significant.

It should also be noted that the comparison is sensitive to data dispersion. Since dispersion is a measure of variance, as the data becomes more varied it is more difficult to separate small differences between data sets. While the geometric means may be different by a factor of 2 or 3 with only 4 points on each side, if the dispersion is on the order of 2 or 3 the test may not recognize a significant difference. Similarly, if the means are effectively identical and one side is highly dispersed the test may not see the two data sets as equivalent.

In this report, pooled variance comparisons are used as a screening tool to flag changes. To maintain some control over the comparison, "before" data is limited to a period spanning four years before the change and after data is limited to a period of 4 years after the change. The extent of the change is indicated by the R-value as explained above. If there is insufficient data to make a comparison, a null value or '-' is indicated. Even if a statistically significant change is indicated, it may be small relative to the scaling factor precision standard and not warrant reanalysis of scaling factors. For the purpose of this report, it merely, highlights changes for consideration. Furthermore, it may miss an important change because of lack of data or because of the quality of the data compared. In these case visual examination of the charts may provide more insight.

Trend Test

To test for a trend, scaling factor log values are regressed according to date. A linear regression analysis is performed along with a slope = 0 test to see if the scaling factor is varying with time.

Analysis Methods

If the null hypothesis (H_0 : $\beta_0=0$) is true, means statistically that the slope cannot be determined to be different from 0 at the 95% level and, effectively, the scaling factor is constant with time.

Given the true slope β_0 , (assumed to equal 0) the calculated slope, b, and variables x and y, the test statistic is defined by

$$t = \frac{b - \beta_0}{s \sqrt{s_{xx}}}$$
(3-9)

where

$$s^{2} = \frac{s_{yy} - b s_{xy}}{n - 1}$$

and

$$S_{xx}^{2} = \sum_{i=1}^{n} (x_{i} - \overline{x})^{2}$$

$$S_{yy}^{2} = \sum_{i=1}^{n} (y_{i} - \overline{y})^{2}$$

$$S_{xy}^{2} = \sum_{i=1}^{n} (x_{i} - \overline{x})(y_{i} - \overline{y})$$

In this case the slope is equivalent to a rate of change of the log value of the slope per day. Given a slope m and one years worth of days (t = 365), the percent change per day can be determined from $(10^{\text{mt}}-1)*100$.

Graphical Analysis

Graphical analysis is useful as a screening tool. The plot is set up with mile stone markers represented by a vertical line through the graph and tagged at the top with a label specific to the change. A key to the labels is provided at the bottom of the graph for easy reference. Individual data points are represented by a solid square. If the value is based on a detection limit, the square appears empty. An individual waste stream can be highlighted within the graph. When highlighted, the particular data points corresponding to the highlighted streams will appear as triangles. In each plot, the scaling factor values are plotted on a log scale against sample date which is printed on a linear scale. The scaling factor data can be normalized. In this case the chart will display the ratios of individual sample points to the median of the first 5 points. The ordinate (y-axis) label is appended the word "norm" to indicate that it is normalized.

By plotting the data in a trend view it is possible to see significant changes in the ratios by visual examination. It doesn't take too much analysis to see that something is going on with the data shown in Figure 3-1, below. In this case Fe-55/Co-60 data is included for four major streams. A large drop is observed following the start of an identified stellite reduction (SR) program. The ratio recovers to original values 10 years later following the introduction of feedwater iron injection (FII). The increase with iron injection seems understandable. The drop following the

start of the stellite reduction campaign is counter to expectations. This leads to new questions about what is actually occurring and might stimulate a search for other events affecting the ratio.



Figure 3-1 Sample Trend Plot Used for Graphical Analysis

Sample Analysis Program

The analysis methods and tools described above form the framework of the 10CFR61 Sample Analysis Program. The program is used in this study to perform the statistical calculations as well as generate the scaling factor charts used to highlight data behavior in the face of particular events.

The program provides the user options to select particular date spans for evaluating scaling factors, forming data comparisons, and examination of trends. The introduction of milestone flags and automated data comparisons before and after the flag was made in support of this report.

4 PLANT WASTE STREAMS

BWR Waste Streams

Reactor Water Cleanup System

The reactor water cleanup system removes impurities from the reactor cooling system. Because coolant enters in liquid form and leaves as steam there is a constant accumulation of non-volatile solids either carried in with the feedwater or being generated through corrosion of fission product production. The reactor water cleanup system includes a powdered resin filter/demineralizer system. Waste is collected as spent powdered resin.

Condenstate Cleanup System

This processes condensed steam from the main condenser system. In most BWRs this is another filter/demineralizer system. A smaller number of plants have instead mixed bed demineralizers. Some solid fission products and TRU carry over with the main steam and are removed with this system. Greater amounts of volatile fission products are carried over. The bulk of these are removed through the condenser vacuum system and collected in the off-gas treatment system.

Radwaste System

More variation is observed in BWR radwaste systems. Original GE designs focused on general use of powdered resin or diatomaceous earth precoated filters. In many cases these systems are in use. In later plants, where more customer input was admitted the systems may include deep bed demineralizers and charcoal filters in conjunction with the precoat filters. In a number of operating plants, the original system has been substituted with a vender supplied liquid waste processing system. These include a combination of processing steps involving varying waste products such as charcoal, zeolites, and specialized resins and additives. Because of these differences, it is difficult to generalize the effects of plant changes without fairly detailed knowledge of the actual processing systems employed.

PWR Waste Streams

(Because of variations in PWR designs differences exist in the primary coolant processing systems. This discussion attempts to be as general as possible.)

Plant Waste Streams

Primary Resins/High Activity Resins

Primary coolant water quality and chemical parameters are maintained through a chemical and volume control system. A letdown flow is maintained to adjust for expansion and contraction during reactor heating and cooling and for removal of accumulated contaminants. The letdown flow is processed through filters and a deep bed demineralizer system before being returned to the reactor.

Low Activity Resins

Low activity resins are generated in the liquid radwaste processing system. This system collects from a variety of sources including equipment drains and floor drains. Original PWR liquid waste processing systems were focused on a evaporation as a corner stone of the system. Few of these systems made it into service and none are being used today. In their place are a range of systems using a combination of processes in series including mixed bed demineralizers, zeolites, charcoal filtration. A few plants are using vendor supplied concentration systems that blend concentrated liquids directly with a waste binder. The by-products of these processes may be mixed for disposal but are usually individually sampled when replaced.

Cartridge Filters

Cartridge filters are used extensively in, mostly Westinghouse, PWR water treatment systems. Because of the nature of contaminant removal with cartridge filters, they can often be regarded as a single stream including letdown filters, fuel pool filters, seal injection filters, boron recycle filters, radwaste processing filters, etc.
5 INDIVIDUAL PLANT ANALYSES

Plant Selection

A number of candidate plants were examined for this evaluation. Factors effecting selection included the actual existence of separable events that may have effected waste stream characterization and scaling factors in particular. Another factor in this selection was continuity of the data base over a sufficient period of time to study the impact. Ultimately, four BWRs and four PWRs were selected for examination. These cover most of the target events identified for study and give a general representation of the data shifts that might be expected with the events.

Milestones Application

It is not always expected that scaling factor changes will be noticed immediately following onset of a change in operations. When a change is introduced it takes some time for the effect to translate through the system. This is particularly true where there are several transport steps for activity from the reactor system to reach the sample stream. In the case where the stream is directly tied to the primary cooling systems (e.g. primary or RWCU resins) samples are collected when the beds are changed out usually during an outage. These samples reflect the previous year's operation. It will take a full cycle longer before samples will be collected that reflected the changed operating condition. Milestones are marked where the change is introduced. Evaluation of the change may be deferred to account for transport delays and for delays in reaching a new equilibrium

A fuel failure event will be observed almost immediately in the reactor coolant via iodine ratios or in the case of BWRs in the condenser air ejector through elevated noble gas activity and changes in noble gas ratios. The impact on scaling factors, if any, is isotope dependent and specific to individual ratios.

In the case of multiple unit facilities, the analysis is limited to Unit 1. It is generally observed that the majority of sample data are referenced to unit 1 or are common to both units. It is often not possible to identify sufficient data specific to second unit to conduct any kind of statistical evaluation. When chemistry changes are introduced, the changes are introduced at or near to the same time in both units.

Streams

Stream selection in individual plants varies according to how an individual plant segregates material, how they sample, sample frequency, and continuity of the data base. Major streams in

a BWR consist of RWCU filter demineralizer resins, radwaste resins, condensate resins, and DAW. For PWRs major waste streams include primary resins, letdown filters, other filters, and DAW. There may be various other streams using a variety of media that are changed out on an irregular schedule, mixed with other material and applied over varying durations. These streams usually may have a spotty sample record or have not been in service long enough to establish a defined operating history.

BWR1

Milestones

Table 5-1 BWR1 Milestones

Designation	Description	Approx. Date	Impact Delay (Est.)
FF	Fuel Failure Event	6/1/1995	
DZO	Depleted Zinc Oxide Injection	12/1/1999	300
PF	Pleated Filter Change	12/1/1997	
NM	Noble Metals Chemical Addition	3/1/2000	

Results

Table 5-2 BWR1 Fuel Failure Impac

Ratio/Stream	RWCU	RW Resins	Condensate	DAW
Sr-90/Co-60	10	1	3	5
Sr-90/Cs-137	10	1	4	10
Pu-239/Co-60	2	1	2	1
Pu-239/Cs-137	2	1	4	3

Ratio/Stream	RWCU	RW Resins	Condensate	DAW
Sr-90/Co-60	1	1	2	1
Sr-90/Cs-137	1	1	1	1
Pu-239/Co-60	1	1	1	2
Pu-239/Cs-137	1	1	1	1

Table 5-3BWR1 Pleated Filter Change Impacts

There was insufficient data following the application of DZO and NMCA to evaluate a change in ratio.

Analysis

The most significant scaling factor shifts occur in the Sr-90/Cs-137 ratio and in the Sr-90 to Co-60 ratio following the fuel failure event. The change is present in other waste streams but is harder to characterize because of the higher dispersions in the ratios in those stream. Figures 5-1 and 5-2 below demonstrate the changes in the Sr-90/Co-60 and Sr-90 to Cs-137 ratios respectively. In both cases the ratios change sharply signifying a relatively large breach in a fuel pin. It is notable that the Sr-90 increases are seen relatively quickly in the RWCU resins.



Figure 5-1 BWR1 Sr-90/Co-60 Reactor Water Cleanup Resins







A lesser shift in the Pu-239/Co-60 ratio is observed, Figure 5-3. Although there is a noticeable increase in the ratio (approximately a factor of 4). The variance test indicates no change. Similar results are observed with other transuranics radionuclides indicating a significant upward shift which is gradually diminishing over several years. Previous investigations have speculated that transport times for transuranic radionuclides from the reactor cooling systems can take 1 to 2 years. This is likely because the highly electropositive ions adhere readily to surfaces and gradually diffuse outward from the reactor.



Figure 5-3 BWR1 Pu-239/Co-60 Reactor Water Cleanup

BWR2

Milestones

Table 5-4 BWR2 Milestones

Designation	Description	Approx. Date	Impact Delay (Est.)
SR	Stellite Reduction	12/1/1990	
FII	Feedwater Iron Injection	8/1/1998	
HWC	Hydrogen Water Chemistry	1/1/1999	

Results

Table 5-5BWR2 Stellite Reduction Impacts

Ratio/Stream	RWC	RW Resins	Condensate	DAW
Sr-90/Co-60	1	1	1	1
Sr-90/Cs-137	-	1	1	1
Pu-239/Co-60	1	1	1	1
Pu-239/Cs-137	-	1	1	1
Fe-55/Co-60	2	5	3	7

Feedwater Iron Injection

Table 5-6 BWR2 Feedwater Iron Injection Impacts

Ratio/Stream	RWC	RW Resins	Condensate	DAW
Sr-90/Co-60	1	1	1	3
Sr-90/Cs-137	-	2	1	1
Pu-239/Co-60	1	1	1	3
Pu-239/Cs-137	-	1	1	1
Fe-55/Co-60	1	2	1	6

Hydrogen Water Chemistry

Table 5-7BWR2 Hydrogen Water Chemistry Impacts

Ratio/Stream	RWC	RW Resins	Condensate	DAW
Sr-90/Co-60	1	1	1	1
Sr-90/Cs-137	1	1	1	1
Pu-239/Co-60	1	1	1	1
Pu-239/Cs-137	1	1	1	1
Fe-55/Co-60	1	1	1	1

Analysis

This plant has a very substantial data base with 76 samples representing the four focus streams. This allows for a relatively complete examination of the operating changes.

Since two of the milestones more directly impact activation products an additional case was run to examine impacts on the Fe-55/Co-60 ratio, refer to Figure 5-4. Both the stellite reduction program and feedwater iron injection showed significant shifts in the Fe-55/Co-60 ratio. In the stellite reduction case the ratio was actually lower than before the start of the program. There is no clear explanation for this behavior. Communication with plant personnel identified no other programmatic changes. If Co-60 was being reduced the ratio would be increasing.

In the case of feedwater iron injection the ratio predictably increases. The largest impact reported is with the DAW stream which indicates a verifiable difference of a factor of 6. However, it is observed that the variance increases in RWCU and condensate following the start of feedwater iron injection which broadens the base of comparison. Discounting some extreme values would indicate that all of the streams included are comparably effected by the changes. Figure 5-7, below isolates the RWCU stream for the period just before and following the start of FW iron injection. By inspection, it appears that there is an upward shift of a factor of 10, but because of variance introduced by outliers the shift cannot be confirmed by statistical test.

Case	Number of Points	Geometric Mean	Dispersion
Overall	7	13.5	5.0
Case After	3	40.6	1.3
Case Before	3	2.57	6.5

Table 5-8BWR2 Fe-55/Co-60 Before and After Comparison

Similar experience is not observed with the Ni-63/Co-60 ratio. Refer to Figure 5-5. No significant impacts are observed on fission product or transuranic ratios to either Cs-137 or Co-60 that can be attributed to the operational changes examined.



Figure 5-4 BWR2 Plant 2 Fe-55/Co-60 Four Streams



Figure 5-5 BWR2 Ni-63/Co-60 Four Streams



Figure 5-6 BWR2 Fe-55/Co-60 RWCU following Start of FII

BWR3

This plant has operated with no significant fuel failures in its operating history. Cesium is usually not picked up in local spectrum analysis and is not used as a basis for scaling. Cesium is reported and tracked in radiochemical samples allowing for the testing of scaling factors. Transuranic concentrations are often below detection limits. The most notable impact on scaling factors occurs with the Sr-90/ Cs-137 ratio following the first chemical decontamination. In this case a statistically significant change of a factor of ten is observed. A scaling factor trend plot for this case is shown in Figure 5-7, below. Most likely this change doesn't have anything to do with the chemical decontamination. More likely, assuming no new fuel failures, fuel contamination was introduced with new fuel.

Milestones

Table 5-9 BWR3 Milestones

Designation	Description	Approx. Date	Impact Delay (Est.)
D1	1 st Chemical Decontamination	4/1/92	
PU	Power Uprate	6/1/94	
D2	2cd Chemical Decontamination	5/1/95	
DZO	Zinc Injection	9/1/1996	
FII	FW Iron Injection	6/1/1996	

Results

Table 5-10BWR3 First Chemical Decontamination Impacts

Ratio/Stream	RWC	RW Resins	Condensate	DAW
Sr-90/Co-60	1	1	1	1
Sr-90/Cs-137	10	1	1	-
Pu-239/Co-60	5	1	-	-
Pu-239/Cs-137	-	-	-	-
Fe-55/Co-60	5	1	1	1

Table 5-11 BWR3 Power Uprate Impacts

Ratio/Stream	RWC	RW Resins	Condensate	DAW
Sr-90/Co-60	5	1	1	-
Sr-90/Cs-137	1	-	1	-
Pu-239/Co-60	1	-	-	-
Pu-239/Cs-137	-	-	-	-
Fe-55/Co-60	1	1	1	1

Table 5-12	
BWR3 Second Chemical Decontamination Impacts	

Ratio/Stream	RWC	RW Resins	Condensate	DAW
Sr-90/Co-60	5	1	1	-
Sr-90/Cs-137	1	-	1	-
Pu-239/Co-60	1	-	-	-
Pu-239/Cs-137	-	-	-	-
Fe-55/Co-60	1	1	1	1

Table 5-13BWR3 Depleted Zinc Oxide Injection Impacts

Ratio/Stream	RWC	RW Resins	Condensate	DAW
Sr-90/Co-60	5	1	1	-
Sr-90/Cs-137	1	-	1	-
Pu-239/Co-60	1	-	-	-
Pu-239/Cs-137	-	-	-	-
Fe-55/Co-60	1	1	1	1

Table 5-14BWR3 Feedwater Iron Injection Impacts

Ratio/Stream	RWC	RW Resins	Condensate	DAW
Sr-90/Co-60	5	1	1	-
Sr-90/Cs-137	6	1	1	-
Pu-239/Co-60	1	-	-	-
Pu-239/Cs-137	-	-	-	-
Fe-55/Co-60	2	1	1	1

Analysis







Other notable changes with this plant were the shifts in the Sr-90/Cs-137 ratio and Sr-90/Co-60 ratio in RWCU following the start of DZO and FII. These two cases are shown in Figures 5-8 and 5-9, below. Since the two changes are so close together in time it isn't possible to distinguish which is driving the shift. However, since the two process work in concert to the same end, they could be viewed as a single milestone. In both cases there is a sustained drop in the scaling factor that is statistically significant at a factor 5. The objective of these processes is to mobilize Co-60 to reduce depositions in crud layers. Since the Co-60 is more mobile in the coolant it is likely that it will be relatively increased in the RWCU filter/demineralizers thereby lowering the scaling factor.

Figures 5-10 and 5-11, provide trends on Fe-55/Co-60 and Ni-36/Co-60 in reactor water clesnup resins. In both cases the scaling factor decrease significantly following the first chemical decontamination. They remain more or less low during the cluster of following changes and begin to recover to their original values. Typically, Fe-55 appears less stable in the later samples.



Figure 5-8 BWR3 Sr-90/Co-60 Reactor Water Cleanup System



Figure 5-9 BWR3 Sr-90/Cs-137 Reactor Water Cleanup System



Figure 5-10 BWR3 Fe-55/Co-60 Reactor Water Cleanup System



Figure 5-11 BWR3 Ni-63/Co-60 Reactor Water Cleanup System

BWR4

Milestones

Table 5-15 BWR4 Milestones

Designation	Description	Approx. Date	Impact Delay (Est.)
PU	Power Uprate	10/1/1996	180
DZ	Depleted Zinc Addition	10/1/1996	180
NM	Noble Metals Chem.Add.	10/1/1998	
HW Hydrogen Water Chemistry		5/1/1997	

Milestone events DZ and PU are not separable since both effects start at the same time. A delay of 180 days is imposed to allow a shift to later sample sets. Data for radwaste/low activity resins was not available.

Results

Table 5-16

BWR4 Power Uprate/Depleted Zinc Addition Impacts

Ratio/Stream	RWC	Condensate	DAW
Sr-90/Co-60	10	1	1
Sr-90/Cs-137	1	1	1
Pu-239/Co-60	1	1	1
Pu-239/Cs-137	1	1	-
Fe-55/Co-60	1	1	1

Ratio/Stream	RWC	Condensate	DAW
Sr-90/Co-60	3	1	1
Sr-90/Cs-137	1	1	1
Pu-239/Co-60	1	2	1
Pu-239/Cs-137	1	1	-
Fe-55/Co-60	1	1	1

Table 5-17BWR4 Noble Metals Chemical Addition Impacts

Table 5-18BWR4 Hydrogen Water Chemistry Impacts

Ratio/Stream	RWC	Condensate	DAW
Sr-90/Co-60	10	1	1
Sr-90/Cs-137	1	1	1
Pu-239/Co-60	1	1	1
Pu-239/Cs-137	1	1	-
Fe-55/Co-60	2	1	1

Analysis

As seen in Figure 5-12, below, the Sr-90/Cs-137 ratio takes a relatively dramatic and sustained drop following the power uprating. This is likely due to fuel changes introduced with uprate. A similar trend is observed with the Sr-90/Co-60 ratio over the same time span.



Figure 5-12 BWR4 RWCU Sr-90/Co-60

Because of the proximity (in time) between the power uprate and the start of HWC it is difficult to separate the effects. However as argued previously, if the primary function of DZO is to mobilize the Co-60, the decrease in the scaling factor is likely due to increasing collection of Co-60 in the stream media.

A factor of 2 shift was seen in the Fe-55/Co60 scaling factor following the start of noble metals addition. This shift can be observed in Figure 5-13 below. The driving force behind this shift is uncertain. Actual values between 1997 and 2000 are more on the order of a factor 10. Noble metals addition is intended to scavenge excess hydrogen ion. At the same time it may be causing a shift in the propensity of iron to form more mobile complexes.



Figure 5-13 BWR4 Fe-55/Co-60 RWCU

PWR1

Data were made available from 1994 through 2002 for this evaluation. Notable events occurring within the data span were small fuel defects occurring on two occasions first in 1994 and again in 1997. Steam generators were replaced in 1996. Hydrogen peroxide is used to control crud buildup in the primary systems. Following steam generator replacement flow was increased to the letdown demineralizers to further limit activity buildup in the primary. There was insufficient data prior to the 1994 fuel failure to evaluate the impact of the event so it was excluded. For the purpose of this evaluation waste streams were separated into 4 groups, resins, filters, miscellaneous solids (solids), and DAW. Low activity filters and resins from secondary waste streams were excluded from the evaluation.

Milestones

Table 5-19 PWR1 Milestones

Designation	Description	Approx. Date	Impact Delay (Est.)
SG	Steam Generator Replacement	6/1/1996	180
FF	Small Fuel Defect	10/1/1996	180

Results

Table 5-20PWR1 Steam Generator Replacement Impacts

Ratio/Stream	Primary Resins	Primary Filters	Process Resins	DAW
Sr-90/Co-60	-	1	1	-
Sr-90/Cs-137	-	1	1	-
Pu-239/Co-60	-	1	1	-
Pu-239/Cs-137	-	1	1	-
Fe-55/Co-60	-	1	1	-

Table 5-21 PWR1 Small Fuel Defect

Ratio/Stream	Primary Resins	Primary Filters	Process Resins	DAW
Sr-90/Co-60	-	1	1	-
Sr-90/Cs-137	-	1	1	-
Pu-239/Co-60	-	1	1	-
Pu-239/Cs-137	-	1	1	-
Fe-55/Co-60	-	1	2	-

Analysis

As seen from the tables above, the statistical tests didn't yield evidence of significant changes in the scaling factor values. A closer look at the data, does show some increased variability in the filter scaling factors following the two events. The Pu-239/Co-60 (shown in Figure 5-14, below) ratio for primary filters shows more scatter about a year after the fuel failure. This is consistent with estimated transport times for plutonium of 1 to 2 years from the reactor to external systems. To large extent this scatter is due to the varying types of filters including primary seal filter, reactor water filter, and letdown filter. It can also be postulated that additional plutonium if present would be taken out in the lead filters and the effect would be diminished as liquids are passed through various process stages. Interestingly, the variation has almost disappeared in the most resent sample set. A similar observation is made for Sr90/Co-60. In this case the scatter occurs more rapidly. This effect is not visible in primary resins as shown in Figure 5-15 below.

As noted above, steps were taken concurrent with the steam generator replacement to reduce activity buildup within the steam generator and primary piping. Over the long term, without

significant changes in materials used in the primary, it would be expected that ratios would return to earlier values. Over the short term following steam generator replacement and the relatively long outage that accompanies it Co-60 concentrations would be reduced. This should be visible in the scaling factor data. Figures 5-16 and 5-17 show Fe-55/Co-60 ratios for primary filters and resins respectively. In both cases the ratios are up slightly following the steam generator replacement.

The point must also be made that the expected result of the chemistry changes accompanying steam generator is to reduce the Co-60 concentration which would tend to increase the scaling factors. The expected result of the fuel defect is to increase the fission product and TRU concentrations which would also increase scaling factors. As a result, it makes it more difficult to speculate as to which result is actually being seen. It may suffice to note that all of the observed changes are relatively small and of limited duration. By the time the sample cycle catches up, with operations, the spike if any has already passed and the effect is spread through the waste volume.



Figure 5-14 PWR1 Pu-239/Co-60 Primary Filters





Figure 5-15 PWR1 Pu-239/Co-60 Primary Resins

[Fe-55]/[Co-60] - Norm



Figure 5-16 PWR1 Fe-55/Co-60 Primary Filters





Figure 5-17 PWR 1 Fe-55/Co-60 Resins

PWR2

Milestones

Table 5-22 PWR2 Milestones

Designation	Description	Approx. Date	Impact Delay (Est.)
SG Steam Generator Replacement		6/1/2000	
UFC Ultrasonic		1/1/2002	

Results

There was insufficient data following the introduction of ultrasonic fuel cleaning for numerical evaluation. Observable changes related to steam generator replacement are listed below.

Ratio/Stream	Primary Resins	Primary Filters	Process Resins	DAW
Sr-90/Co-60	5	1	1	1
Sr-90/Cs-137	2	2	1	-
Pu-239/Co-60	2	1	1	-
Pu-239/Cs-137	2	1	4	-
Fe-55/Co-60	1	1	1	1

Table 5-23PWR2 Steam Generator Replacement Impacts

Analysis

The maximum impact is observed Sr-90/Co-60 for primary resins. A trend plot for this ratio is presented in Figure 18 below. This change is pretty readily observable. Again, the increase in the ratio signifies a decrease in the Co-60 concentration more than an increase in the Sr-90 concentration. In fact, as can be observed in the following Figure 5-19, Sr-90 actually decreases relative to Cs-137. It should be noted that a number of modifications were made with the new steam generator to contribute to an overall reduction in Co-60 concentrations and scale buildup within the steam generators. The scaling factor shift is a result of conscious changes to reduce primary system activity and part of an ongoing ALARA program.



Figure 5-18 PWR2 Sr-90/Co-60 Primary Resins



Figure 5-19 PWR2 Sr-90/Cs-137 Primary Resins

Given the sharp impact on the Sr/Co ratio it's notable that Fe/Co ratio is fundamentally unaffected. Refer to Figure 5-20.



Figure 5-20 PWR2 Fe-55/Co-60 Primary Resins

PWR3

Milestones

Table 5-24 PWR3 Milestones

Designation	Description	Approx.Date	Impact Delay (Est.)
EO	Extended Outage with Steam Generator Replacement	9/1/1997	

Results

Table 5-25 PWR3 Extended Outage/Steam Generator Replacement

Ratio/Stream	Primary Resins	Primary Filters	Process Resins	DAW
Sr-90/Co-60	-	1	-	-
Sr-90/Cs-137	-	2	-	-
Pu-239/Co-60	-	1	-	-
Pu-239/Cs-137	-	1	-	-
Fe-55/Co-60	1	1	2	1

Analysis

A definite shift is seen in the Fe-55/Co-60 (Figure 5-21) ratio following steam generator replacement. Data before and after show a change o about a factor of 10. The effect is obscured by one data point occurring in 1999. If this entry is viewed as an anomaly, then a true factor of 10 shift would have occurred. Including the data point, reduces the statistically evaluated shift of about a factor of 2. The change does seem to be stable as evidenced by later sample points.

[Fe-55]/[Co-60] - Norm



Figure 5-21 PWR3 F55/Co60 Primary Resins

PWR4

Because of its early implementation of zinc injection, this plant provides a unique opportunity to examine the impact. In this plant the waste streams are separated into four groups: primary resins, radwaste mixed resins, DAW, and filters. A sixth waste stream charcoal filter media was also identified from the data but disappears as a separated stream after about 1996. No distinction of plant unit was observed in the resin samples. Most of the filter sample were referenced to originate in one or another of the units however no consistent differences were observed.

Milestones

Table 5-26 PWR4 Milestones

Designation	Description	Approx.Date	Impact Delay (Est.)
DZ1	Depleted Zinc Injection	6/1/1994	

Results

Table 5-27 PWR4 Zinc Injection

Ratio/Stream	Primary Resins	Filters	Radwaste Resins	DAW
Sr-90/Co-60	2	1	1	1
Sr-90/Cs-137	1	-	1	1
Pu-239/Co-60	1	1	1	1
Pu-239/Cs-137	1	-	2	1
Fe-55/Co-60	1	1	1	2

Analysis

The Sr-90/Co-60 ratio showed a statistically significant in primary resins following the introduction of zinc injection. A similar shift can be observed in the Sr-90/Cs-137 trend. In this case a possible outlier in the 1994 sample group precluded statistical confirmation . Trend charts for these ratios are shown in Figures 5-22 and 5-23, below. Inspection of the Sr-90/Cs-137 chart in Figure 5-23 indicates that the change could be substantially greater than a factor of 2 as in the cobalt case. In both cases, the ratio is observed to be on a relatively continuous drift in the upward direction with drops in values occurring at various points. These discontinuities in the trend serve to emphasize the difference. In spite of this, however, there is a change in the value through the course of the period. However, because of the rising trend, it cannot be concluded that the difference before and after initiation of zinc injection has anything to do with the changing ratios. In both cases the ratios begin to decline in 1998 and "level out". The fact that both scaling factors are rising implies increasing Sr-90 relative concentrations. Without evidence of any event surrounding the fuel, it is most likely that the increasing ratios reflect improving demineralizer performance. The leveling off and decline may be more representative of the impact of zinc injection.

[Sr-90]/[Co-60] - Norm



Figure 5-22 PWR4 Sr-90/Co-60 Primary Resins

[Sr-90]/[Cs-137] - Norm



DZ1 - Zinc Injection(Unit 1)

Figure 5-23 PWR4 Sr-90/Cs-137 Primary Resins

6 CONSTANT SCALING FACTORS

A significant number of operating power plants have experienced relatively uniform scaling factor values determined through their sampling programs. This, in concert with a continuing desire to reduce the costs of maintaining, the program has led to more attention given to defining constant values for some ratios aimed at reducing the number and types of radiochemistry procedures performed on each sample. Effectively, each radionuclide requested to be examined by the laboratory invokes a separate procedure and a separate cost factor. By reducing the number of analyses, the overall cost per sample can be reduced.

Candidate Ratios

Ratios most consistent in the laboratory results are those between transuranic (TRU) radionuclides. While some plants scale TRU radionuclides individually to a key radionuclide, most derive ratios between the TRU, usually keyed to Pu-239. A separate scaling factor then is only needed to determine Pu-239 from one of the key gammas – usually Co-60. Because of the consistency of the TRU ratios over time and between waste streams, it would not be necessary to rerun the full analysis for TRU with each new sample. Furthermore if the TRU key were switched from Pu-239 to the more abundant beta-gamma Pu-241, alpha analysis could be eliminated.

Next on the scale of consistency are the ratios between long-lived activated corrosion products and Co-60. Fe-55 and Ni-63 are produced in predictable proportions to Co-60. Because of their chemical similarity to Co-60 (all are of close atomic weight and from the same group in the periodic table), their behavior in various waste streams is similar and proportions remain consistent throughout all streams. More variability is seen in the Fe-55/Co-60 ratio mainly as a result variations in oxidation potentials but also as a result of decay. The delay time depends on the process stage during which the sample is drawn and the duration of use of the collection medium. Never-the-less, the Fe-55/Co-60 ratio is generally consistent within individual streams.

A third group comprises the fission products Tc-99 and I-129. These radionuclides are usually scaled to Cs-137. Both are low energy beta emitters with very long half-lives. As a result special effort is required in their determination by the laboratory including chemical separation and long count times. They are most often reported as detection limits. Consequently, little actual correlation is observed. Since chemical separation is conducted before measurement is attempted, the detection limit is effectively the same for all waste streams. Put another way, the concentration is independent of the key nuclide concentration. Because of their long half-lives, these radionuclides are singled out in 10CFR20 and must be included on disposal manifests even if the detection limit is reported. Most plants continue to define scaling factors for these radionuclides based on Cs-137. Because the detection limit based concentration is the same for

all key values, the scaling factor for high activity streams is low and those for low activity streams are high. Consequently, the I-129 and Tc-99 activities reported for low activity streams are over-stated by 5 or 6 orders of magnitude, while those in high activity streams may be over-stated by only 2 or 3 orders of magnitude. Figure 6-1 shows at typical scaling factor scatter plot for I-129/Cs-137 including all waste streams. Values reported as detection limits as shown as open squares or triangles. Triangles in this plot corresponding to the last sample from a particular stream. No change in behavior is observed with respect to values reported as real. For Cs-137 concentrations ranging from 10^{-6} to $100 \,\mu$ Ci/gm, the I-129 concentration is bounded by approximately 10^{-4} . The scaling factor defined by these data is ~3E-3. A similar result is obtained for Tc-99/Cs-137 as shown in Figure 6-2. In both cases, there is neither correlation nor trend.



Figure 6-1 I-129/Cs-137 Typical Scatter



Figure 6-2 Tc-99/Cs-137 Typical Scatter

Criteria

No criteria have been established for defining constant scaling factors. In the cases of activation products and transuranic ratios, existing radiochemistry data provide an adequate basis for defining these values within the general characterization objective to ensure quantification within a factor of 10 in accordance with the Branch Technical Position on Classification. This condition can be met if the data cover a sufficient operating period and the variance or dispersion of the data is sufficiently small. The dispersion is a measure of how well the value is known. A dispersion of 5 indicates that the true value can be a factor of 5 times higher or 5 times lower than what we regard as the scaling factor. In either case, application of the central value will achieve the general objection of prediction within a factor of 10. For the candidate scaling factors the dispersions are generally observed to be much lower depending on the particular ratio and the extent of segregation of streams.

While it cannot be argued that the past predicts the future, a history of constancy builds confidence that future measurement will be bounded by the cumulative dispersion.

Selection of Values

Currently, all scaling factors are re-evaluated periodically based on cycle length (12-18 months). A rolling average or geometric mean value is determined and used as the current scaling factor. To assure viability of the constant scaling factor, the evaluation should include sufficient data to develop reasonable confidence intervals around the scaling factor and define a scaling factor within the interval. The confidence intervals define a range such that there is a high probability that a new measurement will fall within the interval. If the confidence interval is sufficiently tight it will ensure that the true value will not be under or over estimated to the extent that the general precision objective of plus or minus a factor of 10 is compromised. This would be achieved if the range is limited to a factor of 10 from the high value to the low value. In fact, for the candidate scaling factors ranges as low as a factor of 2 are achievable at the 95% confidence level. Since the objective is precision, rather than conservatism, defining an enveloping value would systematically overestimate and increase the chance of exceeding a factor of 10. Confidence limits themselves are probably inadequate for defining action limits for scaling factors. The confidence intervals defined based on log comparisons are too tight and leave as many as half of the values outside of the intervals. Having established a sufficiently low dispersion, say a factor of 2 or 3, practical intervals can be defined by multiplying and dividing by the dispersion. As upper and lower bounds on the observed values, the intervals should contain at least 90% of the values included in the assessment. Then, when future measurements are compared, they will likely be within the intervals which will avoid an unnecessary reassessment and lot of new samples.

Expectation of Constancy

An expectation of constancy is established by examining the behavior of the scaling factor over time. Specifically, if the ratio has been at or near the same value for several years prior, it would be expected that future measurements will reinforce past experience. A test for constancy can be achieved when the scaling factor ratios are regressed by reference date and subjected to a slope=0 test. A regression by time will identify any trends in the data. A slope = 0 test determines at a defined confidence level if the slope is statistically the same as 0. If the null hypothesis H_0 : m = 0 is true, this demonstrates that the scaling factor is not changing with time. It is possible for some ratios that small trends are embedded in the values. In these cases, the trend should be used to project to a new sampling date and demonstrate that the scaling factor if left unchanged will remain within the precision intervals established for the values used.

Obviously, the longer the period that constancy can be observed, the more powerful the argument that the value is not changing or that it is changing at an observable and defendable rate. Confidence in the projected value can be established by stable operating conditions, absence of trends, strong correlation of data, and low dispersion.

Constant Scaling Factor Example

Figure 6-3, below, provides a good example of a candidate scaling factor for applying a constant value. The figure represents the same ratio for five different waste streams collected from a 2 unit plant over a twelve year period. The overall dispersion is a factor of 2. Statistical analysis

results shown in Table 6-1 indicate a maximum factor of 2 difference between individual streams. Variation from the overall geometric mean is only about 40 %



Figure 6-3 Ni-63/Co-60 Examples

Stream	All	CHAR	DAW	FIL	PMR	RMR
Count	77	6	13	24	15	19
GeomMn	0.989	0.888	0.647	0.8	1.374	1.377
Dispersion	2.121	2.03	1.666	2.358	1.513	2.17
CHAR	1		1	1	1	1
DAW	1			1	2	2
FIL	1				2	2
PMR	1					1
RMR	1					
Trend (%yr)	17	0	14	33	0	0

Table 6-1 Constant Scaling Factor Waste Stream Comparison

The table includes a comparison of each stream with the group as well as comparisons between streams. The second column indicates the statistical difference factors between the overall grouping with each stream included. None of streams are distinguishable within the grouping. Differences of a factor of 2 exist between the two resin streams and DAW and filters. There is a small overall trend in the data which means that it is trending upward at a rate of 17% per year. At this rate of change it would take approximately 15 years for the scaling factor to change by a factor of 10. Given a dispersion of 2.12 and an overall scaling factor of 0.989 the bounding values can be defined as 2.1 and 0.47. Confirmatory measurements taken outside of this range should be investigated. The investigation should start with a cross check on the laboratory.

7 SUMMARY AND CONCLUSIONS

PWR/BWR Differences

In general, the PWRs included in the evaluation provided much greater challenges to generalizing the effects of changes. There were several reasons for this. Firstly, waste stream groupings are more diversified. While all of the PWRs included here are Westinghouse plants, there can be several different streams comprising filters, as well as high and low activity resins. In some cases material from different streams are sampled individually prior to packaging. In others, a composite sample may be taken from packaged material. Cartridge filters samples may be taken from the same stream each time or from one of a variety of other streams. BWRs, on the other hand, have very similar reactor water cleanup systems. All use filters pre-coated with powdered resins with process rates based on percentage of feedwater rates. Because no chemicals are added to control reactivity, the water is basically pure providing for more uniform processing conditions.

Secondly, there are fewer incidences of common milestones and probably more incidences of individual milestones that are not particularly marked. In the case of BWRs, most confronted the issue of $IGSCC^5$ around the same time and began hydrogen water chemistry and other remedial measures. Zinc injection to control surface activity concentrations has been practiced since the early 1990s and there is sufficient data surrounding the initiation of the process to support evaluation. PWRs are only just beginning to experiment with zinc injection and even the motivation for doing it varies. Because of the late start in introducing this process, there isn't sufficient operating history to determine whether or not there has been a change in scaling factor values or to quantify the impact. This effectively precludes a general conclusion.

Thirdly, among the PWRs that have undergone steam generator replacement, a number of other changes have been introduced in connection with it. These made include changes in steam generator materials, passivation of the steam generator bowl cladding to reduce corrosion rates and scale build up, replacement of adjacent primary piping, full or partial system decontaminations, and initiation of chemistry changes on restart. Because these changes occur concurrently, the effects on one change relative to another can't be separated. Along with that, there is usually an extensive shutdown period to provide for replacement, this disrupts any equilibrium that might have been established prior to the maintenance and it may be a full cycle after replacement before new samples are collected that can be applied to the impact.

⁵ IGSCC – Inter-granular Stress Corrosion Cracking

Scaling Factor Process

Samples for scaling factor evaluations are collected on an annual basis. In PWRs, primary resin samples and some filter samples can only be collected at the end of a cycle when the process media are replaced, the sampling period may be extended for as long as two years. In these cases, there is often no effort even to define scaling factors. Rather, the radiochemistry results are used directly as a representative spectrum. There is no stream of similarly composed packages during the course of the cycle to that would require scaling factors. BWRs, on the other hand, are continuously discarding precoat material from the reactor water cleanup system on roughly a bi-weekly basis. Sampling each batch would incur unacceptable personnel exposures as well as be inordinately expensive. For BWR reactor water cleanup resins, it makes more sense to define scaling factors and trend data. In any case, if there is a transient event that would impact scaling factors, much of the impact may have passed by before the next sample is drawn.

To the extent that it can be observed in the data, shifts in scaling factor values can be tied to events including fuel failures, coolant chemistry changes, fuel design changes, major equipment changes. However, as seen in the above analyses, these shifts are almost always within the precision of the overall process and would not necessarily warrant a re-analysis of the scaling factors. Furthermore, the events tend to be transient with the scaling factors reverting more or less to an equilibrium value.

Conclusions

- Changes in plant conditions do have the capacity to shift critical scaling factor values.
- The overall precision of the process for deriving scaling factors envelopes the impacts observed.
- Scaling factor shifts caused by these changes are relatively small in contrast to the overall dispersion of the data and would not in themselves necessitate a reassessment of the scaling factors.
- Operators should be cognizant of these changes but they need not alter sampling schedules around them.
- Candidate ratios for constant scaling factors are generally unaffected by operational changes.
- Constant scaling factor evaluations should include a rigorous evaluation of trends and should establish action envelopes for values.
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