

## **IMPACTS OF OPERATIONAL CHANGES ON LLW SCALING FACTORS**

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### **ABSTRACT**

Since the implementation of 10CFR61 in 1984, US nuclear power plants have adopted, as a routine, sampling and radiochemical analysis for the determination of difficult to measure radionuclides limited by 10CFR61 for shallow land disposal. Samples are collected on, approximately, an annual basis and the results are used in the determination of scaling factors. NRC guidance on the characterization and classification identifies the need to update the sample basis for the scaling factors more frequently if there are significant changes in plant conditions that might potentially change the scaling factors by more than a factor of 10.

A number of examinations of industry experience with sampling and scaling factor development have been completed. In these studies data is assembled as a single database and ratios are analyzed to determine overall averages broken down by streams. All of the conditions that might give rise to changes in the scaling factors for an individual plant are averaged over in the composite grouping. This gives little guidance on how to handle local changes in plant conditions.

Since 1984, several significant changes in plant water chemistry management have been introduced. These include for BWRs, zinc injection to limit the incorporating of Co-60 in the corrosion layer, hydrogen water chemistry to displace oxygen to limit stress corrosion. There have also been changes in PWR water chemistry such as varying lithium concentrations to control formation of corrosion layers. In addition, there have been both BWR and PWRs that have experienced fuel failure episodes, 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.

This work was undertaken for the Electric Power Research Institute. The paper will focus on an explanation of the project, analytical methods, and provide an overview of some of the findings.

### **INTRODUCTION**

With the 10CFR61 rulemaking in 1983, the NRC issued two Branch Technical Positions (BTPs) to provide guidance to nuclear power plant operators on the implementation of the new rules. These BTPs addressed waste form and waste characterization. Radiological characterization included the determination and reporting of activities of a number of difficult to measure radionuclides specifically identified in 10CFR61. This wasn't a new idea. Prior to 10CFR61, low level waste generators were required to report concentrations of transuranic radionuclides in LLW to assure that disposal restrictions previously established for AEC/DOE LLW disposal

sites were observed. Sampling and radiochemical analysis of waste samples was already practiced prior to 10CFR61 primarily to verify TRU concentrations were less than 10 nCi/gm. What 10CFR61 did was expand the list to include several pure beta and low energy photon emitting radionuclides. The BTP on waste characterization noted the impracticality of sampling and measuring all LLW and allowed the use of "scaling factors" based on representative sampling of separable waste streams. The scaling factors are effectively correlations between a difficult to measure (DTM) radionuclide, such as Sr-90, and a 'key' radionuclide that could be readily measured using equipment and methods available to the generator. The Branch Technical Position said that a scaling factor approach to determining the content of the DTM radionuclides was permissible as long as there was programmatic review and updating of the correlations on an annual basis or more often if there was a significant change in plant conditions that might effect the correlations. In accordance with the BTP, scaling factors must be derived on a plant specific basis. It is not sufficient to use industry data.

As a direct result of 10CFR61, operating plants undertook more aggressive sampling programs. Independent streams of LLW were identified, sampling procedures were written, contracts were written with radiochemistry laboratories, and a number of samples began to be sent each year to the laboratories.

## **RELATED EPRI STUDIES**

Several studies and initiatives have been undertaken by EPRI since the introduction of 10CFR61. Two milestone studies were completed in 1985 and 1987. These were: EPRI NP-4037, Radionuclides Correlations in Low-Level Radwaste and EPRI NP-5077, "Updated Scaling Factors in Low Level Radwaste". In both of these studies, sample data were collected from power plants and correlations were calculated for major waste streams including high activity and low activity resins, evaporator concentrates, filters, and dry active waste. Both studies provide perspective on the range of data collected, which scaling factors were most promising in terms of best correlation, and provided baseline data for plants in undergoing startup and unable to develop correlations from their own streams. An additional data collection exercise was completed in 1991 in support of the development of the RADSOURCE<sup>1</sup> computer program. At this point the database was expanded to more than 3000 samples. Correlations were again calculated but this time for a finer breakdown of waste streams. In this examination of scaling factor data, an effort was made to examine the impacts on scaling factors from fuel defects. It was observed that most of the ratios would not be effected by defects since individual radionuclide release rates to reactor coolant were similarly effected. This would be the case with iodine and cesium where both species would proportionately pass through the defect. Alternatively if the release rates were independent of the fuel performance, such as in the case of cobalt and nickel, scaling factors would not be expected to change. While this study was never independently published, it formed a large part of the basis for the 1998, EPRI "Low Level Waste Characterization Guidelines" report. (EPRI TR-107201).

One observation that evolved from these studies was that ratios of certain radionuclides were particularly well behaved. In particular, nickel and iron to cobalt ratios were generally consistent between waste streams and didn't vary much over long time spans. There were differences

between plants but these differences were generally consistent with the dominant materials used in the primary system.

Ratios between transuranic radionuclides (TRU) were also nearly constant between waste streams. These also were observed to not change much over time. How transuranic scaling factors are derived depends on plant practice. Some plants scale all of the transuranics of interest individually to a key gamma radionuclides, either Co-60, Cs-137, or Ce-144. Others, develop one of the transuranics (usually Pu-239) as key to the others, and then evaluate only the key radionuclide correlation with one of the key gamma radionuclides. TRU/TRU ratios show long term constancy as well as constancy between waste streams.

Another observation that evolved from these studies was that certain other radionuclides, in particular I-129 and Tc-99, were particularly difficult to measure (PDTM). While positive results were reported, there was little correlation with available key gamma. Most of the reported values stated concentrations at the limits of detection and were not true concentrations at all. It is probably true for all of the 3000 attempts to measure I-129 in LLW, it was never successfully measured by radiochemical methods. Judgment is reserved on the failure rate of the Tc-99 measurements. All that was sure was that activities of these two radionuclides were systematically over reported by several orders of magnitude. In conjunction with the development of the RADOURCE program, controlled experiments were run in a number of power plants using resin columns to collect samples from primary coolant. These samples were then analyzed by Battelle using mass spectrometry to determine isotopic composition<sup>2</sup>. The measurements confirmed what was already known. Radiochemical methods were not adequate for determining I-129 and Tc-99.

## **DRIVE TOWARD CONSTANT SCALING FACTORS**

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 satisfactory correlation with Co-60 or Cs-137, the trend, 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 radionuclides for which new sample data will contribute meaningful new information. These are Sr-90 and Pu-239<sup>3</sup> that can be effectively scaled to one of the key gammas.

Since 10CFR61 sample collection started, there has been a relatively dramatic improvement in fuel performance in the nuclear power plants. Few plants have had recent fuel failures. As a result, it has become increasingly impractical to use Cs-137 as a key radionuclide since it is often 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 the scaled radionuclide.

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 scaling factors.

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. 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

## **PLANT CHANGES POTENTIALLY EFFECTING SCALING FACTORS**

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:

### **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 occupational 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.

### **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 was more pronounced in plants with deep bed demineralizer systems that were 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**

Lithium is used in PWRs in much the same manner as zinc is used in BWRs. Since PWRs used a substantial amount of Inconel, there is a major concern with amount of nickel that precipitates on core surfaces. Addition of LiOH, reduces the release rates and the deposition Co-58 which is a major activation product of Ni-58. Co-58 contributes significantly to occupational exposures. Over the longer term, controlled lithium concentrations should reduce the corrosion rate of Inconel and thus the release of Co-58 and Co-60. It does not impact release rates from previously deposited scale so the effect on scaling factors should be gradual.

### **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 used have taken place 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 at 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 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.

## ANALYTICAL METHODS

The purpose of this examination is to assess if changes in plant conditions are of sufficient impact to effect changes in scaling factors and necessitate additional sampling. The primary tools used in the evaluation of scaling factors and changes are highlighted below.

### Geometric Mean and Dispersion

Geometric mean (GM) and dispersion (GD) have been the principal methods for evaluation of scaling factor data. The geometric mean (GM) is defined as the  $n^{\text{th}}$  root of the product of  $n$  values. It is usually calculated by the antilog of the average logarithmic ratio. A function to calculate this value is available in Excel (geomean()). If you want to perform the calculation manually, the formula is provided in Equation (1).

$$GM = \log^{-1} \left( \frac{\sum_{i=1}^n \log \left( \frac{y_i}{x_i} \right)}{n} \right) \quad (\text{Eq. 1})$$

Where:  $y$  = Scaled Nuclide  
 $x$  = Key Nuclide

The GM is effectively an estimate of the median value. It is more robust than a simple average in that outliers have less impact on the outcome.

The dispersion, reported extensively in EPRI studies, can be interpreted as a range of difference in the scaling factor ratios used in the GM calculation. A dispersion of 2 would indicate "average" variation by a factor of 2 above and below the GM. To assure prediction within a factor of 10 a  $2\text{-}\sigma$  dispersion admits variation only within a factor of 5. The dispersion is the antilog the variance calculated from the log ratios. The variance is the square root of the standard deviation. A formula for calculating the dispersion is provided in equation (2):

$$\text{GeometricDispersion} = \text{antilog}(\sqrt{S^2}) \quad (\text{Eq. 2})$$

where the standard deviation ( $S^2$ ) is defined using the logarithm ratios.

### Trend Analysis

Trend analysis is performed using linear regression methods. The scaling factor ratios are plotted on a time scale and the regression is performed on the ratio versus time. The slope of the regression can be equated to the rate of change. Using real data, it is unlikely that a slope of 0, indicating no trend, will be calculated. A statistical test can be performed to determine within confidence limits if the slope is zero.

The pertinent equations are presented below:

The test statistic is defined by:

$$t = \frac{b - \beta_0}{S\sqrt{S_{xx}}} \quad (\text{Eq. 3})$$

where  $b$  is the slope estimated by linear regression and  $\beta_0$  is the hypothetical real slope. When used to test for trends, the hypothetical real slope is equal to 0. Other parameters are defined by:

$$S^2 = \frac{S_{yy} - bS_{xy}}{n-1} \quad (\text{Eq.4})$$

And

$$S_{xx}^2 = \sum_{i=1}^n (x_i - \bar{x})^2, S_{yy}^2 = \sum_{i=1}^n (y_i - \bar{y})^2, \text{ and } S_{xy}^2 = \sum_{i=1}^n (x_i - \bar{x})(y_i - \bar{y}) \quad (\text{Eq. 5})$$

### Pooled Variance

Pooled variance is a technique for testing whether two data sets are equivalent (i.e. from the same population). Conceivably, if there is no discernable difference in the data, you could combine and treat it as one data set. A warning is in order in that more internal variation there is in the two data sets, the more likely they are to appear statistically equivalent. You have low confidence in the true value of either data grouping. The data are the same in that you can't draw any conclusion from either set.

Pooled variance is calculated to justify averaging between varying streams. It is applied to individual ratios since not all ratios behave similarly across varying streams. The pooled variance calculation compares data sets on the basis of their variance and degrees of freedom to determine if the data sets are statistically the same.

$$S_b^2 = \frac{(n_1 - 1)s_1^2 + (n_2 - 1)s_2^2}{(n_1 + n_2 - 2)} \quad (\text{Eq. 6})$$

The test statistic used for the comparison is given by:

$$t = \frac{|\mu_1 - \mu_2|}{S_b \sqrt{\frac{1}{n_1} + \frac{1}{n_2}}} \quad (\text{Eq. 7})$$

where:

- $\mu_1$  = Average of Waste Stream 1
- $\mu_2$  = Average of Waste Stream 2
- $n_1$  = Sample Count for Waste Stream 1
- $n_2$  = Sample Count for Waste Stream 2
- $S_b^2$  = Pooled Variance

The pooled variance as employed in this analysis will be the principal test to determine if a change is significant over longer time frames. That is, it will verify if the data prior to the introduction of the change in plant conditions is significantly different from the data collected after the event.

### Graphical Analysis

To support the analysis a graphical depiction of the data was developed to show the scaling factor ratio trended over the collection period. Superimposed on the graph are important milestone events that could impact the scaling factors as well as the plant operating history. A sample graph is shown in Fig. 1.



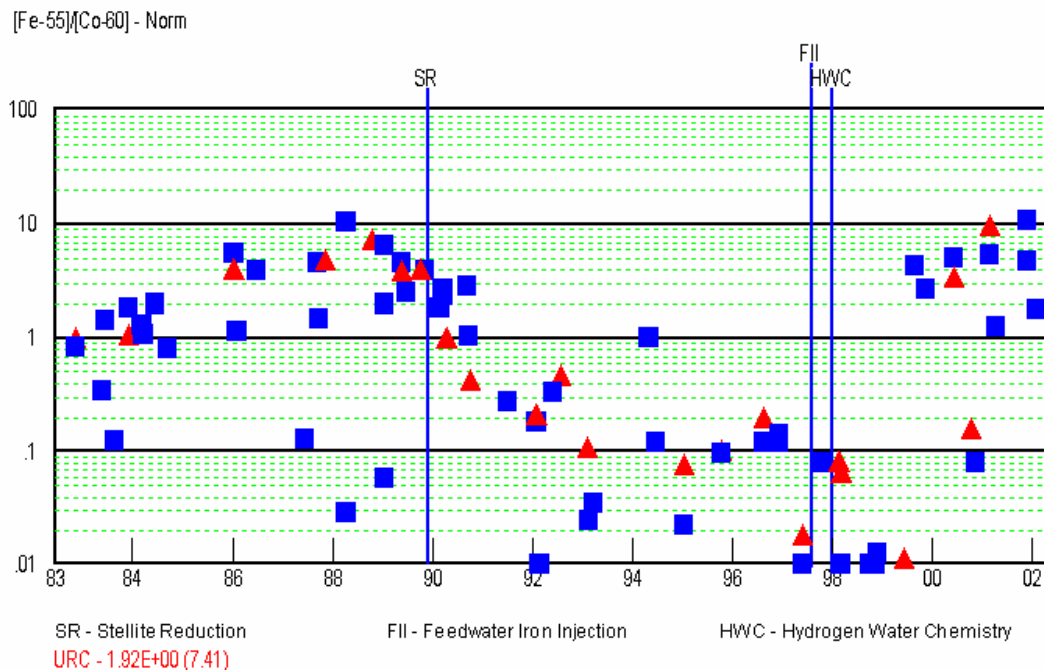


Fig. 1. Scaling Factor Trend Plot with Milestones and Operating History.

In Fig. 1, the milestones marked are:

- SR – Stellite Reduction
- FII - Feedwater Iron Injection
- HWC – Hydrogen Water Chemistry

## 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 being given to defining constant values for some ratios aimed at reducing the number and types of radiochemistry analyses conducted on each sample. Effectively, each radionuclide requested to be examined by the laboratory invokes a separate analysis and a separate cost factor. By reducing the number of analyses, the overall cost per sample can be reduced.

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 objective 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.

Figure 2, 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 %.

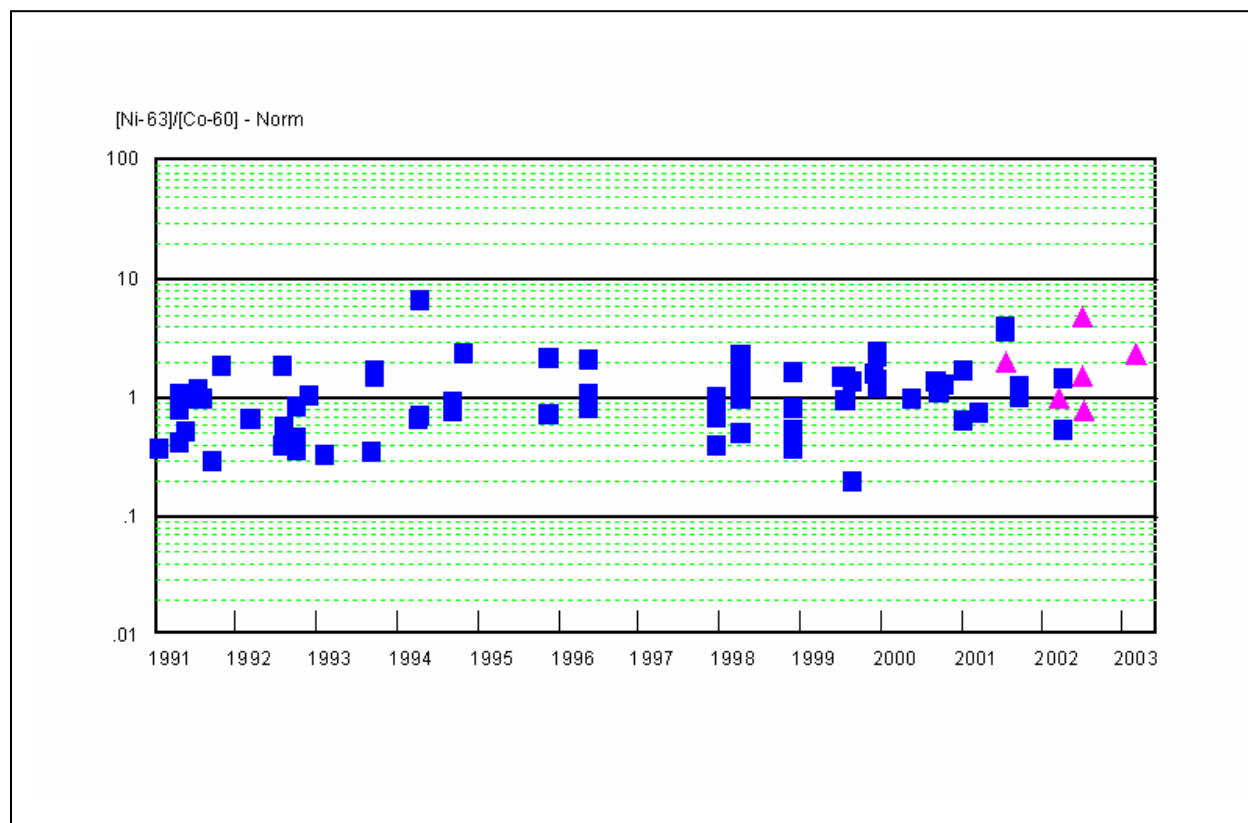


Fig. 2 Constant scaling factor example (Ni-63/Co-60)

## RESULTS AND CONCLUSIONS

Examination of scaling factor impacts requires fairly detailed knowledge of what is occurring in the plant while the scaling factor data is being collected. From the example in Fig. 3. A fuel failure event is indicated followed by more than a factor 10 of increase in the Sr-90/Co-60 scaling factor. In the years following the event, the scaling factor remains high but appears to be trending downward. It is relatively unaffected by subsequent plant operating changes.

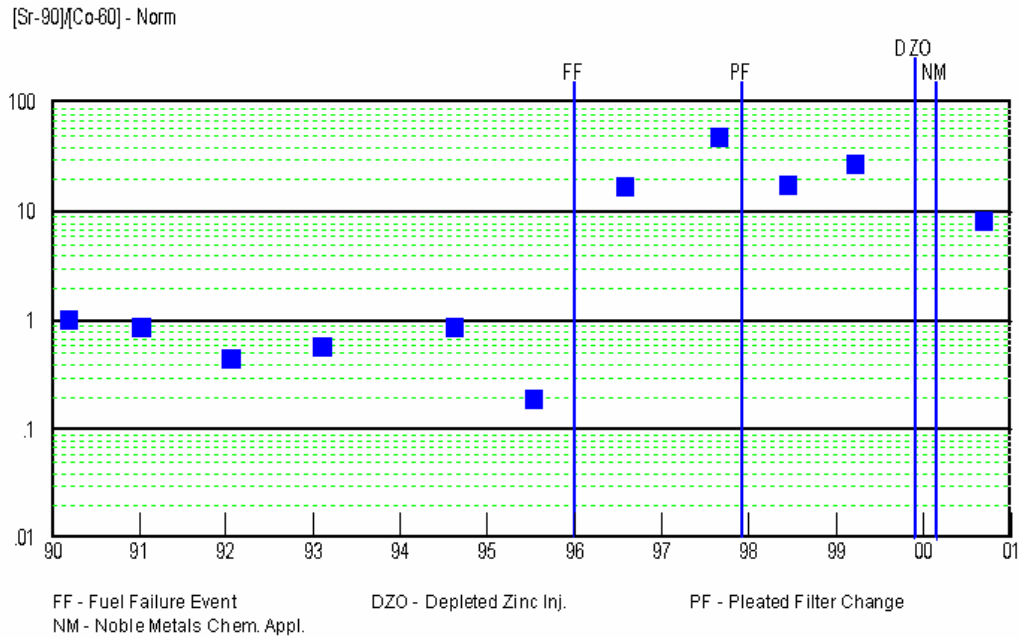


Fig. 3 Impact of fuel failure event

The second example presented here (Fig. 4) demonstrates the shift in the Sr-90/Co-60 ratio in RWCU following the start of zinc oxide injection and feedwater iron injection. 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. 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.

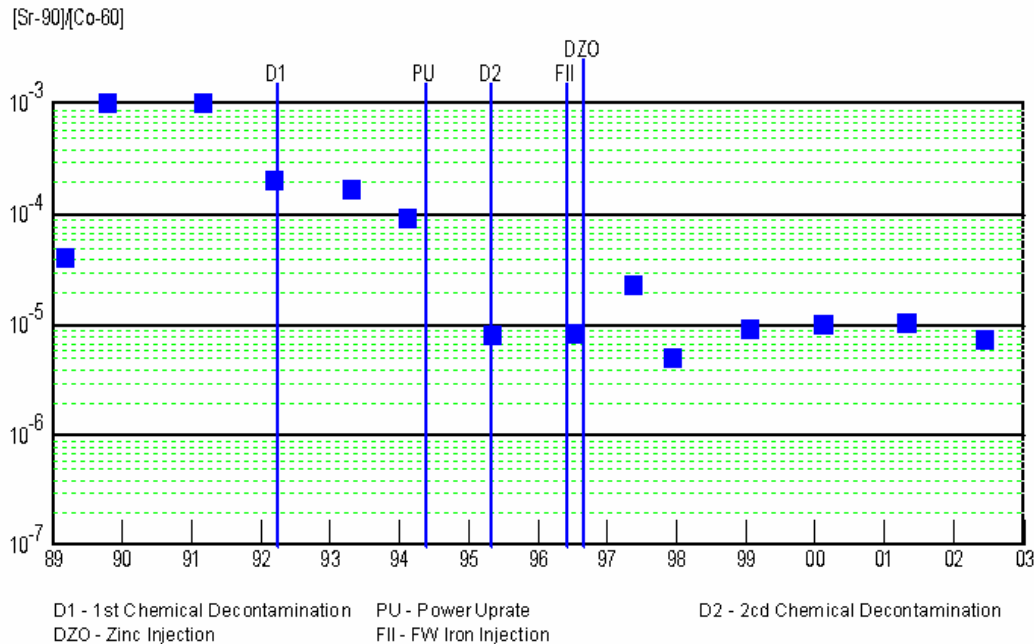


Fig. 4 Scaling factor reduction events

An important overall consideration in this study is whether or not a particular database can support examination. To understand the impact of a change in plant conditions, it's necessary to know when the change occurred and the plant operating conditions surrounding the change. In the example shown, the plant experienced stable operation during the entire period. The data base is well developed with a strong sample record for all of the major waste streams including sufficient data for statistical tests and trend analysis. Owing to the manner in which characterization is performed at some plants less emphasis is placed on the overall database and it can be problematic to attempt to reconstruct the sample record. Hopefully, the results of this effort will provide valuable information to all plants confronting changes potentially effecting characterization efforts.

## 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.

## REFERENCES

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- 1 Developed by Jene Vance, the RADSOURCE program predicted scaling factors for fission product radionuclides from reactor coolant concentrations of radioiodine isotopes. RADSOURCE was developed for EPRI based on the ISOSCALE program developed by Jene Vance.
- 2 D.E. Robertson, et. al., "Concentrations and Behavior of  $^{129}\text{I}$ ,  $^{99}\text{Tc}$ , and  $^{14}\text{C}$  in Low Level Radioactive Wastes from Commercial Nuclear Power Stations, WM'91, Vol 2. pp.287-294
- 3 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.