Building Context for Radioactive Waste Characterization – 14040

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ABSTRACT

Radiological characterization of radioactive waste is required to demonstrate conformance with Federal and State regulations and disposal site license criteria. The Nuclear Regulatory Commission has published guidance for radiological waste characterization that includes an expectation of accuracy. The guidance specifically identifies accuracy as the regulatory objective, i.e. over-estimating waste activity is just as unacceptable as under-estimating waste activity. Most waste generators depend on sample data to perform characterization. How we use this data to best effect however, depends not only on the results from samples that we analyze but also on knowledge of how, and under what conditions the waste is generated and our expectations of what the results should be. Simple sample and measurement data may not be enough in complex situations to develop confidence in the results. Building that confidence requires that we understand the process that creates the radioisotopes, the processes we use to collect samples as well as the processes used to analyze the samples and the potential sources of error associated with each. Data without context does not establish any measurable confidence. Industry research and regulatory guidance point to a number of methods that can be used to build context within which one can establish confidence in waste characterization data. This paper will explore ways to build context and confidence in radioactive waste sample data. While the paper focuses on nuclear power plant wastes, the concepts presented are generally applicable to the overall process of radiological characterization.

INTRODUCTION

What is characterization? Webster's dictionary defines it as the act of characterizing. The word 'characterizing' is defined as: to describe the character or quality of [something]. A 'character' is one of the attributes or features that make up and distinguish an individual [or thing]. The word 'describe' is to represent by a figure, model or picture. (1) Characterization is therefore the description of the inter-related conditions in which something exists or occurs. It is the process to establish coherence between an observation with the environment and the setting in which it exists. When we talk about radiological characterization in the context of waste disposal, we are talking about the process of describing the conditions, environment and processes that result in a particular collection of material that is intended for disposal as radioactive waste. The process is built through repeated observations, measurements, and performance tracking.

Radiological characterization is required to demonstrate conformance with disposal site conditions. These conditions are developed from performance assessments of the site which include the local environmental conditions, facility design and expected waste form, activity and isotopic distribution. The initial conditions and limitations for low-level radioactive waste disposal in the United States were developed in NUREG-0782 (2) and codified in Title 10 to the Code of Federal Regulations, Part 61 (3). The result was a waste classification system with a list of radio-isotopes and their associated concentrations (by volume and mass) that are considered suitable for shallow land disposal under a defined set of circumstances. The list of radio-isotopes includes some that are readily and easily measured and quantified and many that are difficult to measure.

METHODS AND REGULATORY PERSPECTIVE

Radiological characterization can be accomplished through a number of methods including radiochemical analysis of samples, gamma spectrum analysis with correlations to non-gamma radionuclides, calculations of production and removal factors or some combinations of methods. You can rely on individual samples without corroboration, but that requires a truly representative sample. You can't do correlations without radiochemical analysis of samples. You can perform gross gamma analyses without correlations to non-gamma radionuclides but that may miss radionuclides important to the disposal site performance. You can calculate activity based on production and removal mechanisms but you need to reasonably understand the process. The methods are not mutually exclusive and none of these really stand completely on their own. Each method also has limitations and a set of assumptions that must be made with regards to the data. Understanding the results requires comprehension of the processes that create, transport and remove activity in the system. If you don't have an expectation of the results, then your ability to express confidence in the data is limited as is your ability to recognize change.

The Nuclear Regulatory Commission (NRC) identifies 4 basic methods that can be used to comply with the disposal requirements in 10 CFR Part 61. These are described in their Branch Technical Position on Waste Classification (BTP 1983) (4). They are:

- Source or Inventory Control/Process Knowledge.
- Direct measurement.
- Gross Radioactivity (dose to activity).
- Correlations – Scaling Factors (Derived from Direct Measurements).

Most characterization activities applied in nuclear power plants are some combination of these methods.

The NRC expects licensees with complicated waste generation processes (such as nuclear reactors) to use any and all methods necessary to determine accurate activity and waste classification. This expectation is specifically stated in the BTP 1983. The NRC realized however, that accuracy in measuring and reporting activity may be difficult for some waste types and forms. The guidance states that licensees use "reasonable" efforts to ensure realistic representation of activity and defined a "reasonable target" for accuracy as within a factor of 10 of actual concentrations. (4)

Implicit in this expectation of accuracy is that the licensee has to have some knowledge of what the actual activity and concentrations are. Without going into a long treatise on statistics and error analysis, defining accuracy requires some knowledge of what result to expect so that bias can be established. Some of the radionuclides that the regulations require for tracking can't be reliably measured using standard radiochemistry. Some are vulnerable to sample handling. The overall process presents challenges that go well beyond a last sample mentality. The NRC recognized these problems and expected an effort to understand and explain process mechanisms as part of the process to demonstrate compliance.

The NRC's expectation of accuracy applies in both directions. Over-reporting of activity is discouraged as much as under-reporting. This is evident in the statements in Information Notice IE 86-20 'Low Level Radioactive Waste Scaling Factors, 10 CFR Part 61' (5). In the IE notice NRC staff documented inspector's observations that utility waste programs showed poor correlation

between generic radionuclide concentration data and actual radionuclide sample data. Discrepancies greater than a factor of 10 were noted and NRC concluded that the practices could lead to significant over estimates as well as under estimates of actual activity. (5) The IE Notice concluded with four major expectations of utility waste programs:

- Programs should be facility and waste stream specific,
- Method should not unduly over-estimate or under-estimate actual concentrations,
- Concentrations determined from scaling factors should be accurate to within a factor of 10 (also noting that factor of 10 variations are identified as significant and may indicate a possible change or non-compliance but not that they do represent non-compliance)
- Basing activity on a single sample is acceptable – if the sample is representative of the waste as a whole.

The last point is important to further clarify as many licensees rely on the last sample almost exclusively to evaluate a waste container. The text of the IE Notice states 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 combine the latest analysis with those previously obtained to refine the scaling factors currently in use." (5) This implies (rather strongly) that there is a context for making a choice that is based on operating history, sample history for the waste stream or any plant changes that may have occurred and that samples are actually representative of the waste. The preference for using site-specific data and building a history (or context) for evaluating samples is clear. Reliance on the most recent sample was identified as a choice or alternative from the primary expectation of gathering more detailed information over time.

REPRESENTATIVE SAMPLING ISSUES

The question of what constitutes a representative sample becomes the key issue that can be resolved only by building a context within which to evaluate the individual data points. The basic premise of a representative sample is that it is a subset of all the contaminants represented in the same proportions as in the whole population. On the physical side, this requires a well-mixed source, defined and consistent inputs, meticulous sampling technique and statistically relevant sample size. These conditions are almost never fully met in a normal operating environment. Most nuclear plant systems are not designed for good mixing and have multiple inputs to tanks or liners. (6) (7) Most samples consist of <1-10 grams of material used to represent 2,000 to 3,000 kilograms of waste.

Samples must also be representative of the operating period during which the waste was generated. Waste samples sent for laboratory analysis are typically taken at annual intervals. (4) Sometimes they consist of a number of other samples composited over the interval. Radioactive decay is not always accounted for in isotopic ratios. Tank design and operation do not always allow for a complete and even change of material. Therefore tanks may contain mixtures of old and new waste. These factors can significantly affect the distribution of activity from the sample results as compared to any other batch of the waste material.

Decisions for the classification and packaging of LLRW based on non-representative sample data can lead to errors. Under predicting the activity in the waste container can lead to selection of the wrong package for transportation resulting in increased risk to the public and emergency response personnel in case of an incident during transportation and place more activity into the disposal site than evaluated in the performance assessment. Over-predicting activity has the

effect of wasting resources and increasing costs. (5) The use of more restrictive packaging for transportation can increase worker exposures to radiation (packaging and handling multiple containers to fit the smaller Type B packages), increase storage times (due to limited availability of Type B packages) and increase safety risks to workers (due to the need for multiple handling of containers and use of heavier equipment). Disposal site resources are wasted and the site may need to close earlier than intended as the reported activity reaches the performance assessment limits. While the risks of over-predicting seem more palatable from the perspective of public opinion, they may in fact create more problems for society as a whole.

Making the correct decision then requires that each piece of data available is properly evaluated and placed in the proper context so the true nature of the waste can be accurately determined. The results of individual samples are only pieces to be examined and not necessarily the true description of the waste. Data users often look at a concentration obtained from a laboratory as being 'the concentration', without realizing that the number generated by the laboratory is the end point of an entire process, extending from design of the sampling, through collecting, handling, processing, analysis, quality evaluation, and reporting. The U.S. Environmental Protection Agency (EPA) has done studies as part of Superfund clean-up work. In their Soil Sample User's Guide, EPA states that "…data obtained from sampling and analysis are never perfectly representative and accurate, and that the cost of trying to achieve perfect results would be quite high. Consequently, EPA acknowledges that some uncertainty in data must be tolerated, and focuses on controlling the uncertainty which affects decisions based on those data". (8) The recommended approach to evaluate individual sample data is to establish a statistical basis wherein the normal variance of the total population can be described and the results of any single sample can be declared to be representative with a designated degree of confidence. (9) (10)

This is especially problematic in nuclear utility waste streams where on-site analysis capabilities are limited to gross counting and/or gamma spectroscopy. The remaining radionuclides important to classification must therefore be derived from scaling factors based on laboratory analysis of annual samples. Scaling factors thus derived are subject to statistical variances depending on operational issues and representativeness of the sample. (11) When scaling factors are based on the results from a single sample it becomes very difficult to establish the accuracy of the result.

Obtaining truly representative samples of utility waste streams is also problematic. (12) There are many barriers to obtaining a representative sample. Many system designs do not have the capability of sampling until multiple waste types have been transferred to a collection tank. Some systems do not have sampling capabilities until after the waste has been transferred to the disposal container. Mixing capabilities are varied with some sites having no mixing capability at all. Tank designs or operations may not allow complete removal of all material on discharge to a waste container resulting in a mixture of waste from different operating periods and with different radioactive decay periods. (6) The collection of multiple samples for a particular batch of waste is frequently not possible due to dose concerns for the workers and adherence to the regulatory principle of As Low As Reasonably Achievable (ALARA). (6)

Waste sample sizes are frequently limited to less than 500 grams. The sample size sent to an offsite laboratory may be limited by activity and license restrictions to less than 1 gram. (6) On-site samples used for gamma spectroscopy are sometimes less than 1 milligram. The results from such samples cannot be assumed to be truly representative based on size alone.

Many studies have been done to identify methods to determine if a sample is representative. Figure 1 shows the results of sample size on concentration. The study was performed by the U.S Department of Energy in the mid 1970's. The data was derived from 20 replicate aliquots of various mass of a single finely milled (homogenized) sample of 4 kg. The larger sample sizes results in a tighter distribution around the true concentration. The smaller samples result in a wider range of probable results. The data shows that even when you have made efforts to homogenize the sample, variability is inevitable at the laboratory level. (13) Therefore, any decision based on the data is uncertain unless you have some other context to know what the expected value should be.

Figure 1 Effect of Sample Size on Results (http://www.itrcweb.org/ism-1/2_4_1_3_The_effect_of_subsample_mass_on_data_variability.html)

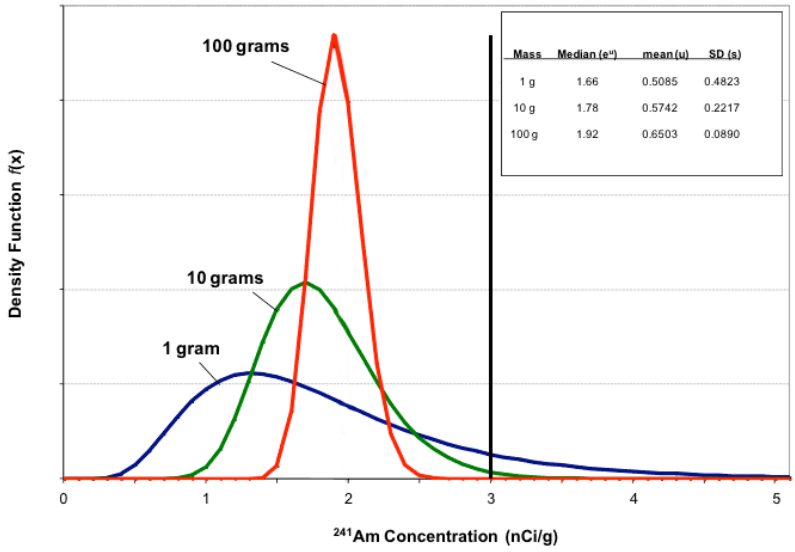


Figure 2-9. Smaller analytical masses contribute to high data variability.
Source: Data from an experimental study on radioactively contaminated soil (Gilbert and Doctor 1985).

The lack of ability to obtain a truly representative sample from utility waste streams defines the issue with respect to characterizing waste. The results of any single sample analysis have very little meaning without some context within which to evaluate the data.

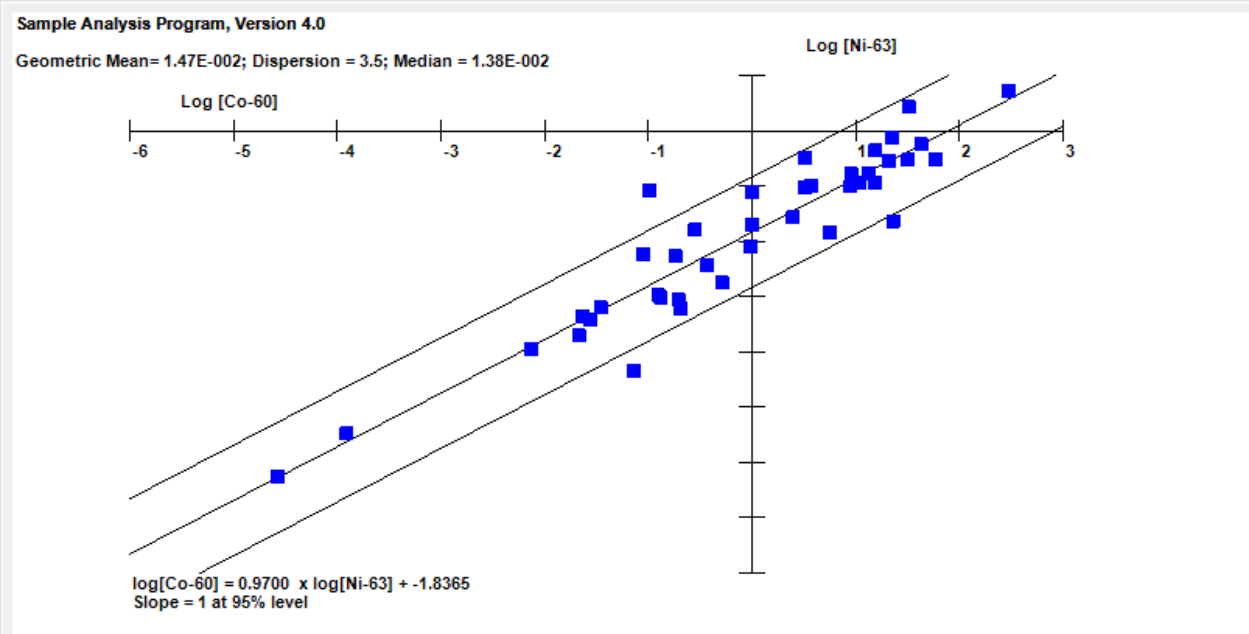
BUILDING CONTEXT

One way to resolve this issue is with larger or a higher number of samples of each waste container. There are a number of problems with this approach namely the additional radiation exposure required to obtain and handle the samples, the lack of laboratory capabilities to accept the increased activity associated with multiple samples, the cost of the additional analyses and the time necessary to perform the analyses on each waste container. The individual sample results would still need to be evaluated together to establish the distribution of the data and develop a measure of the central tendency.

A more realistic approach is to use the sample data collected from multiple waste samples over an extended period of time to build a database. The database can then be used to establish a track record which can be analyzed statistically to develop long-term trending and therefore context for each new piece of data. The characterization developed in this manner would have a measureable confidence associated with it and would provide a more stable basis for determining

waste classification. While not a specific measurement of any single container of waste, the relationships among radionuclides in utility waste are relatively stable over long term operating conditions that are also relatively stable by design. (11)

Figure 2 Typical $^{63}Ni/^{60}Co$ Relationship Over a 9-Year Period



USING STATISTICS

Statistical methods used to evaluate radioactive waste streams are not very different from those used to evaluate any data set and have been used quite often for analyses of radioactive waste. (10) Central tendencies can be computed using simple averages (mean), a geometric mean (log average) and tested to determine how well they describe the overall population by determining the standard deviation and median and performing correlation, regression and pooled variance analyses to test how well the data relates to itself and to other data sets and provide a measure of confidence in the analysis. (11) The Sample Analysis Program (SCAN) developed by DW James Consulting provides a ready platform for analyzing waste sample data using these tests. (14) Specific statistical tests and how they are applied in the SCAN program are discussed below: (15)

Geometric Mean

Geometric mean is often referred to as the log mean average. It is calculated as the antilog of the average log ratio. The geometric mean is used as an estimation of the median. It is preferred in scaling factor calculations because it tends to be less susceptible to outlier influence and is has traditionally been easier to work with than the median. The geometric mean is defined by:

$$Geometric\ Mean = antilog\left(\frac{\sum_{i=1}^{n} log\left({}^{y_i}/_{x_I}\right)}{n}\right)$$

Where:

$y_i = Scaled\ Nuclide$
$x_i = Key\ Nuclide$

Geometric Dispersion
The Geometric Dispersion, as used in this program, is the antilog of the standard deviation, *s*, calculated from the log ratios. The dispersion serves as a measure of variability. It may be interpreted as an average factor of the variation. For example, if the dispersion is equal to 2, then, on average, the data will vary by a factor of 2 from the geometric mean.

$$Geometric\ Dispersion = antilog\sqrt{s^2}$$

$$where\ s^2 = \left(\frac{\left(\sum log\left({}^{y_i}/_{x_I}\right)\right)^2 - \sum log\left({}^{y_i}/_{x_I}\right)^2}{n(n-1)} \right)$$

Where:

$y_i = Scaled\ Nuclide$
$x_i = Key\ Nuclide$

Median
The Median is defined as the middle value in an array of numbers. That is, half of the values lie above the median and half of the values lay below. The median, like the geometric mean, is considered to be a robust estimator because it is not substantially influenced by extreme values or outliers. If the median is found to be significantly different from the geometric mean, you may wish to consider removing outliers or changing your waste stream selection. If the array consists of an even number of values, the median is calculated as the average of the two center values.

Outlier Tests
Outliers are values that are extreme in relation to most of the other data. If statistical outlier checking is used, then whether or not a value is an outlier depends on the amount of overall variation in the data. The greater the variation, the more extreme the outlier value must be. SCAN presently identifies outliers as values exceeding a factor of 10 above or below the median, independent of the data spread. The use of a factor of 10 was selected so that case definitions could be defined consistent with the NRC policy on averaging. That is, values should not be averaged if there is more than a factor of 10 difference from the middle value (i.e. geometric mean or median). Care should be exercised when factor of 10 outliers are removed since the action could move the result in the wrong direction. A better approach to removing outliers is to develop a clearer understanding of the factors that might be influencing the sample results and determine if the number is possible given expected variations in the waste stream. Another option is to narrow the case of the offending values by tightening the date span or cutting the number of streams included in the case.

Outlier Removal
Outliers can be removed if the value exceeds a factor of 10 difference from the geometric mean value. This is statistically arbitrary and does not mean “should”. A true outlier should be discrepant without a physical reason. It is not appropriate to remove data unless there is adequate

justification to do so. Even data identified as a statistical outlier may not be discrepant. Statistical outliers should be investigated to determine if they indicate a change in the waste stream or even an extreme (but possible) value before justifying their removal. The NRC guidance suggests that scaling factors should predict concentrations within a factor of 10 of the actual values. Outlier flagging based on a factor of 10 provides the user with a basis for refining the calculation case (e.g. select a different waste stream grouping or exclude a particular waste stream).

For most nuclide pairs, waste stream differences will not account for an observed difference exceeding a factor of 10. This is true for most Co-60 based scaling of corrosion products and all Pu-239 based scaling factors with exception of Cm-242. In these cases, are usually safely removed since the central value is already well established. For C-14, fission products scaled to cobalt or cesium, more effort is needed in case definition. If you confine your data to the most recent sample periods and waste stream groupings of similar types or individual waste streams (i.e. DAW), you have the best chance of eliminating the outliers. However, you may not be able to eliminate all outliers by case selection. If the median value is significantly different than the geometric mean, it may be a good idea to trim some of the most extreme values.

As an alternative index, the program flags if a point is a statistical outlier using a robust estimator based on deviations from the median. The program calculates the median absolute deviation (MAD) according to the formula:

$$S = 1.483 \underset{j=1,\ldots,n}{\text{median}} \left| x_j - \underset{i=1,\ldots,j}{\text{median}}(x_i) \right|$$

Where: S = the scale estimator and 1.483 is a correction factor to make the estimator consistent with the usual scale parameter of a normal distribution.

Outliers are identified by the statistic:

$$z_i = \frac{x_i - \boldsymbol{T}}{S} > 2.5$$

Where: **T** is identified as the sample median. (Reference: Handbook of Statistical Methods for Engineers and Scientists, H.M Wadsworth, McGraw-Hill, 1990)

Generally, you can identify outliers by observing the scaling factor plot. If the point lies outside of the factor of 10 boundaries, the program will allow you to treat it as an outlier. If the point also represents a statistical outlier it will also be indicated in the Message Box. Note the more disperse the data, the less likely that a given value will be a statistical outlier. You can find out about the data point by clicking it in the scaling factor plot. The click will generate a message box displaying the sample number, date and waste stream. If the point is identified as an outlier (exceeding a factor of 10 from the geometric mean), the box will also include the outlier index and offer you the opportunity to remove it. Up to ten outliers may be removed from a case. Actually if you have to remove more than a few, then you should probably redefine the case.

Pooled Variance

Pooled variance can be used to check the viability of combining individual waste streams. When Sample Analysis Program constructs a case that includes more than one waste stream it counts

the sample data and calculates the geometric mean and variance for each individual stream as well as for the overall data set. It then compares these results using the method of pooled variances described in Walpole, R.E., Meyers, R.H., *Probability and Statistics For Engineers and Scientists*, Fourth Edition, MacMillan Publishing Company, New York, 1989, pp 246-248. (16)

The pooled variance is calculated from:

$$s_b^2 = \frac{\left((n_{1-1})s_1^2 + (n_2 - 1)s_2^2\right)}{n_1 + n_2 - 2}$$

Where s_b^2= pooled variance, n_1 and n_2 refer to data count for data sets 1 and 2 respectively and s_1^2 and s_2^2 are the related variances. The pooled variance is then used to define a test statistic, t, that can be compared with table values such that:

$$t = \frac{|\mu_1| - |\mu_2| - log(R)}{s_b \sqrt{1/n_1 + 1/n_2}^{1/2}}$$

In this equation μ represents the mean of the respective set of data. The mean and variance are calculated using the data logarithms

Test t is then compared with t/2 where its value corresponds to the student t-value at 0.025 (95% Confidence Level). The value R represents a ratio or a difference factor between the two mean values. If R=1, (log(R) = 0), there is no difference - that is, at the 95% level we cannot say that the ratio of the two means is different than 1 and therefore a 95% confidence that there is no difference between the geometric means given the number of samples compared and the observed variances. The R value may be interpreted as a difference factor. If for a given stream comparison, the R value exceeds 3 it may be necessary to keep the stream separate in the analysis.

Regression Test (Slope Testing)

The Slope test is used in conjunction with linear regression of scaling. This test is useful for two purposes. Firstly, the use of the geometric mean presumes linearity between the scale and key radionuclide at a slope = 1. The NULL hypothesis (H_0:) that "slope = 1" is true at the 95% confidence interval, it is concluded that the slope is not different from 1 and provides reinforcing evidence that use of the geometric mean is valid in the case under study. Conversely, a positive test for “slope = 0” indicates that there is utterly no correlation between the evaluated radionuclide and the key radionuclide. The scaled radionuclide is the same value for all concentrations of the key. In this case a valid scaling factor cannot be defined and another key should be selected if available or a constant concentration should be used.

The equations for the comparison are given below:

The test statistic is defined by:

$$t = \frac{b - \beta_0}{s\sqrt{S_{xx}}}$$

where *b* is the regression slope and β_0 is the hypothetical real slope. Other parameters are defined by:

$$s = \frac{S_{yy} - bS_{xx}}{n - 1}$$

And:

$$S_{xx} = \sum_{i=0}^{n} (x_i - \overline{x})^2$$

$$S_{yy} = \sum_{i=0}^{n} (y_i - \overline{y})^2$$

$$S_{xy} = \sum_{i=0}^{n} (x_i - \overline{x})(y_i - \overline{y})$$

Trends

Trending is designed to identify changes over time and accommodate several fuel cycles. Trends should be displayed by plotting the selected ratio normalized to a reference starting ratio. If the starting ratio is representative and the data is not changing, the normalized ratio should scatter around a value of 1 for the entire span of dates.

Trend Test (Slope = Zero)

The Trend Test is performed by checking to determine if the trend slope over time is statistically the same as zero. Regression analysis is used to calculate the slope. The ratio of the slope to the standard deviation of the slope is compared with the student t value corresponding to the 95% confidence level. If the ratio is less than the corresponding t value, the NULL hypothesis (H_0:) that "slope = 0" is true at the 95% confidence interval, it is concluded that there is no trend. If the NULL hypothesis is false, the regression slope is reported on the log plot.

SELECTING RADIOISOTOPES FOR ANALYSIS

The selection of radioisotopes necessary to characterize LLW is based on the requirements in regulations, specifically Title 10, Code of Federal Regulations, Part 61 (10CFR61) (3). These in turn are derived from a series of performance assessments done for typical but generic facility types in different regions of the U.S.. (2) The specific list of isotopes is as follows: (3)

Table 1 10CFR61.55 Table 1

Radionuclide	Concentration curies per cubic meter
C-14	8
C-14 in activated metal	80
Ni-59 in activated metal	220
Nb-94 in activated metal	0.2
Tc-99	3
I-129	0.08
Alpha emitting transuranic nuclides with half-life greater than 5 years	[1]100

Radionuclide	Concentration curies per cubic meter
Pu-241	[1]3,500
Cm-242	[1]20,000
[1] Units are nanocuries per gram	

Table 2 10CFR61.55 Table 2

Radionuclide	Concentration, curies per cubic meter		
	Col.1	Col. 2	Col. 3
Total of all nuclides with less than 5-year half life	700	(1)	(1)
H-3	40	(1)	(1)
Co-60	700	(1)	(1)
Ni-63	3.5	70	700
Ni-63 in activated metal	35	700	7,000
Sr-90	0.04	750	7,000
Cs-137	1	44	4,600
(1) There are no limits established for these radionuclides in Class B or C wastes. Practical considerations such as the effects of external radiation and internal heat generation on transportation, handling and disposal will limit the concentrations for these wastes. These wastes shall be Class B unless the concentrations of other nuclides in Table 2 determine the waste to be Class C independent of these nuclides.			

Typical isotopes found in U.S. utility waste include ^{54}Mn, ^{60}Co, ^{65}Zn and ^{137}Cs. All of these isotopes emit gamma rays (17) and are relatively easy to detect, identify and quantify using gamma spectroscopy methods. However, results from gamma spectroscopy analysis still need to be scrutinized carefully for data that does not make sense. Some typical examples are identifications of short, half-life radionuclides that can no longer be present simply due to the length of time between sample collection and analysis, misidentification or mis-quantification of isotopes due to interference, in adequate accounting for background radiation, non-detection of isotopes due to masking by Compton Scattering, missed or inadequately resolved peaks. Most of these issues can be resolved through an understanding of the context in which the waste was sampled. There should always be an expectation of the isotopes that should be present and those that should not, based on known production mechanisms. Anomalous results need to be investigated and, if not corroborated, eliminated from the analysis. The primary gamma isotopes that make for relatively reliable key-radionuclides

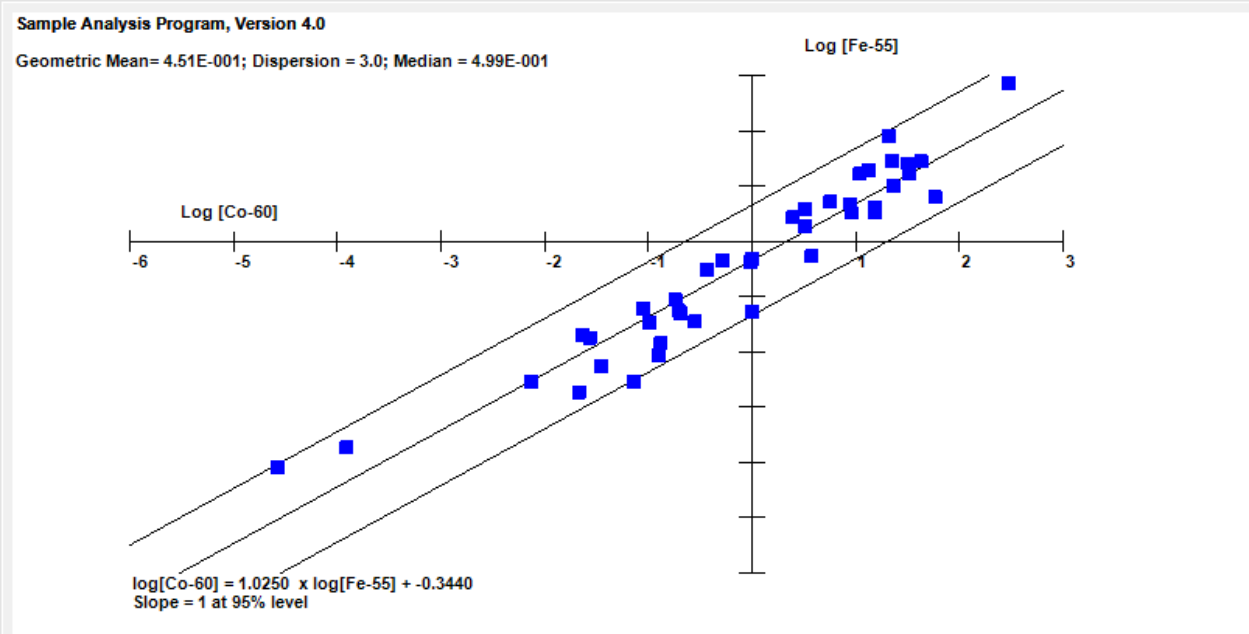
The remaining radionuclides are typically determined though laboratory analysis using chemical separation and other counting techniques and can be correlated to one of the gamma isotopes for routine quantification. (10)

USE OF SCALING FACTORS

Scaling factors work where the relationships, both physical and chemical, between the key

radionuclide and the scaled radionuclide are similar and consistent. (12) Scaling factor pairs are typically developed based on the production mechanism of the radionuclides, i.e. fission products scaled to other fission products, activated corrosion products to other activated corrosion products, etc. For some pairings this works particularly well. ^{60} Co, ^{55}Fe and ^{63}Ni are both constituents of reactor system metals. They are introduced to the system through corrosion and erosion processes and they are chemically similar so they are removed from reactor coolant by similar processes and methods. Long term trending of ^{55}Fe/^{60}Co and ^{63}Ni/^{60}Co show solid and consistent relationships as seen in Figure 2 (above) and Figure 3.

Figure 3 Typical ^{55}Fe/^{60}Co Relationship Over a 9-Year Period



Other radionuclides such as ^{90}Sr and trans-uranic radionuclides (TRU) can also be reasonably scaled to ^{60}Co despite different production mechanisms as they more closely share transport properties and generally have a constant production rate. Examples are shown in Figure 4 and Figure 5.

Figure 4 Typical ^{90}Sr/^{60}Co Relationship Over a 9-Year Period

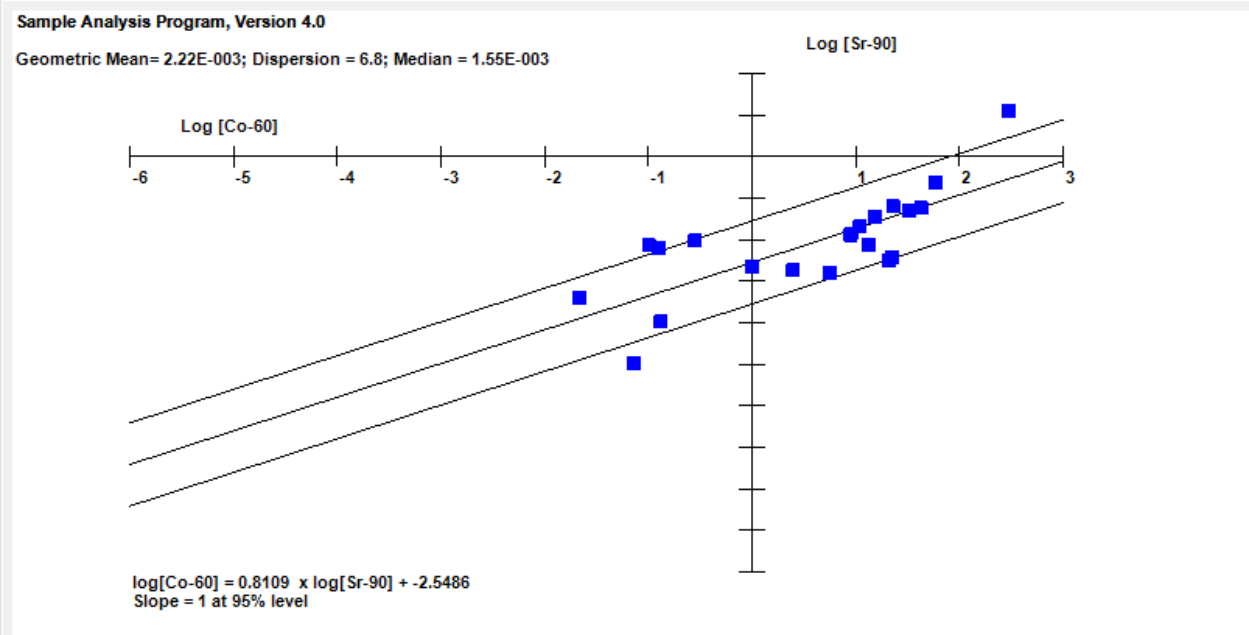
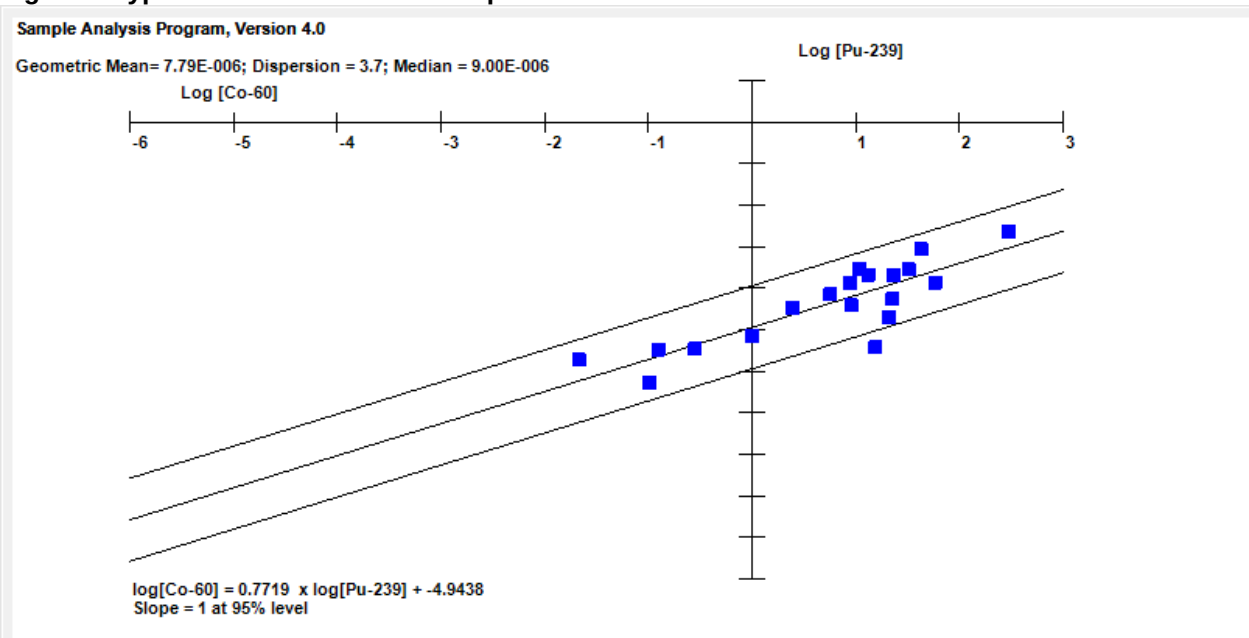


Figure 5 Typical ^{239}Pu/^{60}Co Relationship Over a 9-Year Period



^{90}Sr and TRU's may also be scaled to ^{137}Cs however the relationship may not hold well due to differences in production mechanisms and nuclide transport and removal mechanisms that are specific to ^{137}Cs. (10) TRU's can also be reliably scaled to ^{144}Ce however ^{144}Ce is difficult to measure and is not routinely observed in reactors with good (or even reasonably good) fuel performance. (10)

Some radionuclides do not correlate well with any key radionuclide. Problematic among these are ^{3}H, ^{14}C, ^{99}Tc and ^{129}I which are required for characterization under 10CFR61 and for reporting under 10CFR20 Appendix G. (3) (18) Both ^{3}H and ^{14}C in U.S. utility reactor processed waste

streams are primarily produced from reactor coolant. They therefore have very little in common with either activation or fission produced radionuclides and exhibit nearly constant concentrations over several orders of magnitude changes in the key radionuclides as seen in Figure 6 and Figure 7. ^{14}C is also un-reliably measured in laboratory analyses of utility waste streams at the typical analysis limit used. Sample data shown in yellow in Figure 7 represent laboratory results reported as the lower limit of detection (LLD).

Figure 6 Typical $^{3}H/^{60}Co$ Relationship Over a 9-Year Period

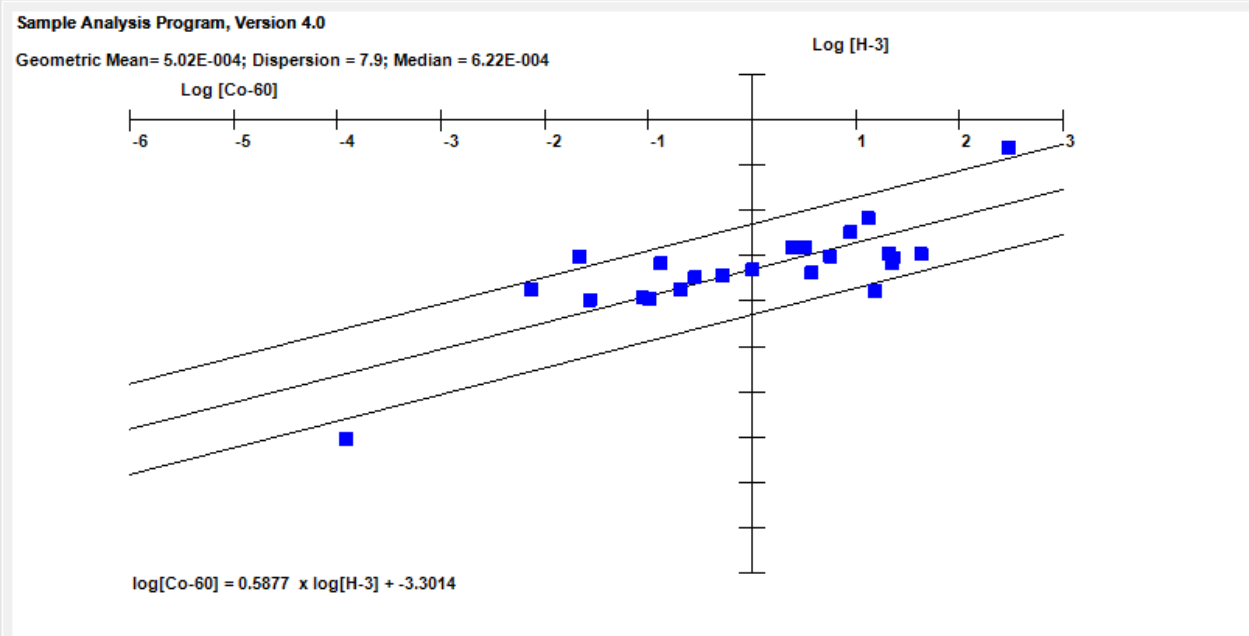
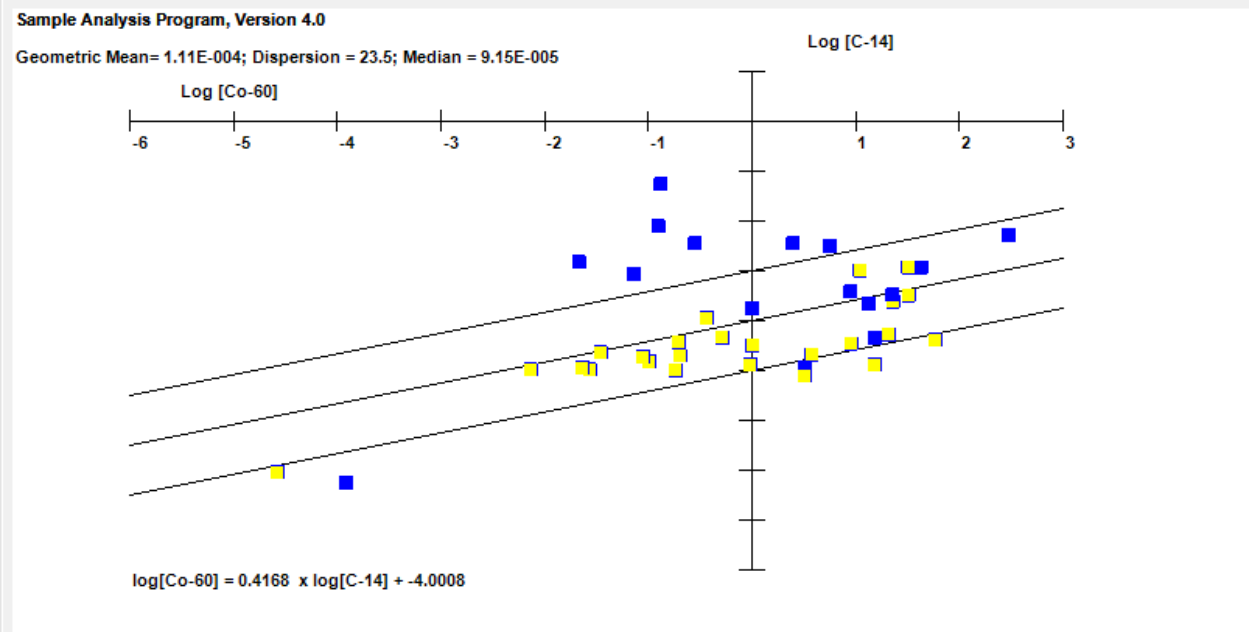


Figure 7 Typical $^{14}C/^{60}Co$ Relationship Over a 9-Year Period



Similar detection issues are present for ^{99}Tc and ^{129}I as shown in Figure 8 and Figure 9. They are not typically identified in waste samples above laboratory LLD's.

Figure 8 Typical ^{99}Tc/^{60}Co Relationship Over a 9-Year Period

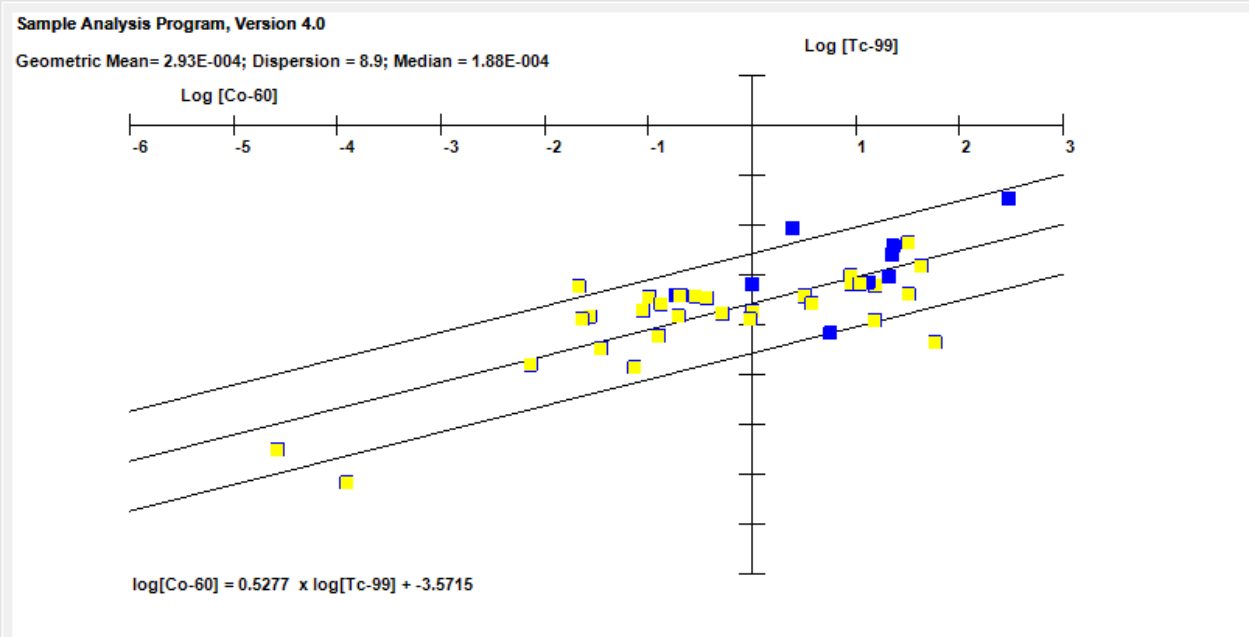
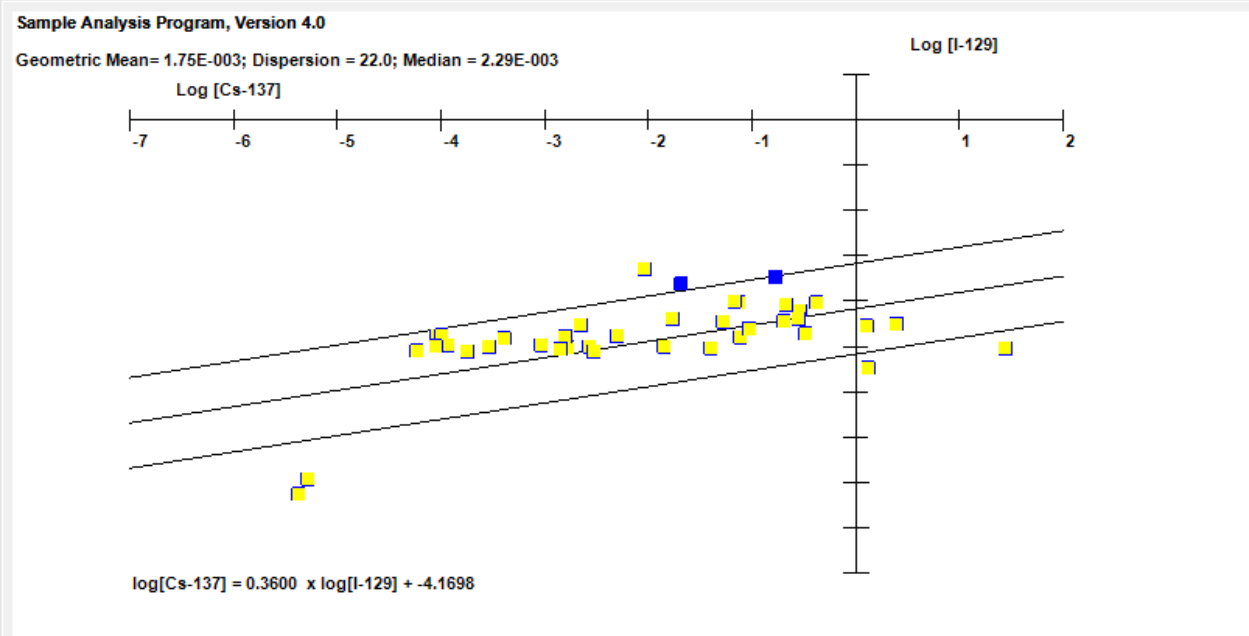


Figure 9 Typical ^{129}I/^{137}Cs Relationship Over a 9-Year Period



ALTERNATIVES TO SCALING FACTORS FROM WASTE SAMPLES

Sometimes waste sample data isn't enough to accurately determine radionuclide activity in the waste. Not all plant systems allow for easy or clear segregation of waste streams or for consistent sampling. Waste stream sampling is also after the fact showing what's been happening in the

reactor as opposed to what is happening. Monitoring reactor coolant data, which is routinely collected for operational purposes, allows for prediction of waste activities and management of waste as it is being generated, i.e. before it is loaded into a waste container. This kind of process knowledge can be used to actively manage waste classification and minimize (if not avoid) Class B/C waste disposal costs.

Reactor coolant is sampled every day for full gamma spectrum analysis. There is ample opportunity to build confidence in plant performance as it applies to estimating radionuclide releases. Concentrations observed in the reactor coolant relate directly to the production rate and removal factors in the reactor system. Some radionuclides such as ^{137}Cs can be monitored directly and efficiently since the sample is uniform and homogeneous. Other radionuclides can be predicted efficiently through surrogates that are not otherwise detected in waste samples due to short half-life. An example is the use of ^{58}Co to determine ^{63}Ni. Analysis methods have already been developed for estimating release rates for important difficult to measure radionuclides. (19)

CONCLUSION

Radiological characterization of radioactive waste is required to demonstrate conformance with Federal and State regulations and disposal site license criteria. The Nuclear Regulatory Commission has published guidance for radiological waste characterization that includes an expectation of accuracy. The guidance specifically identifies accuracy as the regulatory objective, i.e. over-estimating waste activity is just as unacceptable as under-estimating waste activity. Most waste generators depend on sample data to perform characterization. How we use this data to best effect however, depends not only on the results from samples that we analyze but also on knowledge of how, and under what conditions the waste is generated and our expectations of what the results should be. Simple sample and measurement data may not be enough in complex situations to develop confidence in the results. Building that confidence requires that we understand the process that creates the radioisotopes, the processes we use to collect samples as well as the processes used to analyze the samples and the potential sources of error associated with each. Data without context does not establish any measurable confidence. Industry research and regulatory guidance point to a number of methods that can be used to build context within which one can establish confidence in waste characterization data.

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