DETERMINING THE UNCERTAINTY ASSOCIATED WITH RETROSPECTIVE AIR SAMPLING FOR OPTIMIZATION PURPOSES

Dennis J. Hadlock
Westinghouse Savannah River Company
Building 735-2B
Aiken, SC 29808

ABSTRACT

NUREG 1400 contains an acceptable methodology for determining the uncertainty associated with retrospective air sampling. The method is a fairly simple one in which both the systemic and random uncertainties, usually expressed as a percent error, are propagated using the square root of the sum of the squares. Historically, many people involved in air sampling have focused on the statistical counting error as the deciding factor of overall uncertainty in retrospective air sampling. This paper looks at not only the counting error but also other errors associated with the performance of retrospective air sampling. By placing the various errors in the same units (e.g., percent error) it is possible to determine the overall uncertainty for a specific air sample. In the case of this paper, the overall uncertainty when analyzing a 'typical' air sample for Pu-239 at the 0.10 and 10 Derived Air Concentration (DAC) levels using a gas proportional counter was evaluated. For the examples in this paper it was found that the count error dominated the overall uncertainty at the 0.10 DAC where at the 10 DAC level the counting error, although significant, did not dominate. Once this analysis is performed, it is possible to optimize the overall uncertainty associated with the air sample based on the knowledge of which errors influence the overall uncertainty the most. This optimization process allows resources to be directed where they can do the most good. In the examples used in this paper, it was determined that for the 0.10 DAC sample, a simple increase in the daily background count time from 10 to 30 minutes provided a reduction in overall uncertainty of about 15%.

BACKGROUND

The majority of retrospective air samples (RAS) obtained at the Savannah River Site (SRS) are concerned with the alpha component of the source term. Low level counting of alpha emitters will also provide the largest associated counting error due to the small amount of radioactivity needed to exceed personnel protection trigger levels. This should result in a total error that would bound a similar beta-gamma analysis. As such, this paper will deal only with the error associated with alpha analysis of RAS.

Another likely error is how representative the air sample is to what you intended to sample (e.g., personnel or a specific area). Using an extreme example, placing an air sample upwind of a worker performing airborne radioactivity producing work can introduce an error in the results of several orders of magnitude between what you sampled and what the worker was exposed to.

This type of error can easily dwarf any of the other potential errors in air sampling. Performing a quantitative assessment of this type of error would provide little benefit as the possible results and their affect is obvious. Most Radiological Controls Organizations have specific procedures that provide guidance on obtaining representative samples for both workers and areas. The calculations in this memorandum will assume that the air sample is representative.

DISCUSSION

Errors are normally classified into two groups, systematic and random errors [Bowers 1996]. Systematic errors are errors that occur repeatedly in the same magnitude and can be the result of faulty equipment calibration or bias on the part of the observer. For example, if the calibration of a counting instrument was 10% higher than the calibration source indicated, each measurement with that instrument would have a +10% systematic error.

Random errors are ones that result from variations in judgement, experimental conditions, small disturbances, or intrinsic statistical variations in the quantity measured. The exact time of occurrence of a random error cannot be predicted. Random errors will vary in both directions around the true value and will also vary in magnitude. Random errors are evaluated by using statistical analyses.

When there are two or more errors that affect a measurement, the errors must be combined to give the total error associated with the measurement. This is not a simple addition of errors. The errors must be propagated correctly.

When measured values are combined, the associated errors must be propagated by use of equation 1.0 [NRC 1993].

$$E_T = \sqrt{(E_1)^2 + (E_2)^2 + (E_3)^2 + ... + (E_n)^2} \qquad \text{\{Eq. 1.0\}}$$

Where: E_T = Total error

E_n = the error associated with a given measurement expressed in the same units as the value of the measurement.

It is important to note that the terms under the radical in equation 1.0 must all be in the same units. It is usually easier to ultimately express both systematic and random errors as a percent error (%error) to facilitate propagation of the total error.

Air sampling results, in μCi/cc, are determined using equation 2.0.

$$A = \frac{(ncpm)\left(4.5E - 7\frac{\mu Ci}{dpm}\right)}{(eff)\left(\dot{V}\right)(CF)(t_s)(CE)(SA)} \qquad \text{\{Eq. 2.0\}}$$

Where: A = Airborne radioactivity in µCi/cc.

ncpm = net counts per minute of sample. Calculated by:

[(gross cpm) – (background cpm)]

4.5E-07 = Conversion of µCi to dpm.

eff = Detector efficiency.

$\dot{V}$ = Sample flow-rate.

CF = Conversion factor to convert the units of $\dot{V}$ to cubic centimeters.

t_S = Time of sample.

CE = Collection efficiency.

SA = Self-absorption of 0.8.

In the following, each of the input values to equation 2.0 is listed along with (1) whether or not any error(s) is associated with its value and (2) a description of the possible error(s)

ncpm: Error associated with the counting statistics of radioactivity to a 95% confidence level.

4.5E-07: This value is a constant; therefor no error is associated with it.

eff: Error associated with the instrument efficiency would be that associated with the National Institute of Standards and Technology (NIST) traceable source used in the calibration. There is also additional error associated with the daily source checks.

$\dot{V}$: Rotameters are calibrated to within of ±15% of the standard used. Error associated with the calibration standard and reading the rotameter scale must also be considered.

CF: This value is a constant; therefor no error is associated with it.

t_S: Some small error in the sample time can be expected.

CE: No error is associated with the collection efficiency of approved filter papers using the assumption that the filter paper is not damaged. However, if the filter efficiency is ≥95%, no correction needs to be made [NRC 1992]. This can be accounted for as a likely error.

SA: An error should be assigned to this input value based on measurement uncertainty.

Based on the analysis of the input values, there are six that have an error associated with them. They are: (1) net counts per minute; (2) instrument efficiency; (3) flow measurement; (4) sample time; (5) collection efficiency; and (6) self-absorption. Each is discussed in the sections that follow.

Net Counts per Minute

The error associated with counting statistics is the standard deviation (σ) of the count. The σ associated with any particular counting result is determined by the use of equation 3.0 [Cember 1996].

$$\sigma = \left[\frac{\sqrt{n}}{t}\right] = \left[\sqrt{\left(\frac{n}{t}\right)\left(\frac{1}{t}\right)}\right] = \left[\sqrt{(r)\left(\frac{1}{t}\right)}\right] = \left[\sqrt{\frac{r}{t}}\right] \qquad \{Eq.\ 3.0\}$$

> Where: n = total counts (c)
>   t = count time (min)
>   r = count rate (cpm)

In this case we are interested in subtracting one count from another (gross counts minus background counts) and determining the resulting %error of the ncpm based on the σ value. In addition, the resulting %error is affected by the confidence interval chosen for the instrument. Counting instruments typically have a confidence interval of 95%. This corresponds to 1.96σ [Cember 1996]. This means that for a particular activity result, there is a 95% confidence that the actual activity lies between ±1.96σ of the result. Combining equations 1.0 and 3.0 and taking into account the confidence interval of 95%, equation 4.0 can be used to determine the standard deviation of the ncpm.

$$\sigma_n = 1.96\sqrt{\frac{r_g}{t_g} + \frac{r_b}{t_b}} \qquad \{Eq.\ 4.0\}$$

> Where: σ_n = standard deviation, at 95% confidence, of the ncpm
> r_g = gross counting rate
> t_g = time during which the gross count was made
> r_b = background counting rate
> r_t = time during which the background count was made

For purpose of illustration, two situations will be analyzed. The first will be a gross count rate that will produce a result of about 0.1 Derived Air Concentrations (DAC) Pu-239 (2.0E-13 μCi/cc) and the other a gross count rate that results in about 10.0 DAC Pu-239 (2.0 E-11 μCi/cc). In order to determine these count rates, and the resulting σ values, the following assumptions/inputs are used:

- Background is counted for 10 minutes

- Background count rate is 2 cpm

- Sample count time is 45 minutes for the 0.1 DAC example and 1 minute for the 10 DAC example (the count times would typically vary in order to meet the desired Minimum Detectable Concentration (MDC)).

- Sample flowrate is 4 cfm

- Sample time is 2-hours (total volume of 480 ft^3)

- SA equals 0.8

- Detector efficiency is 35%

Using equation 2.0 and solving for ncpm, 0.1 DAC is 1.7 cpm and 10.0 DAC is 169 cpm. For a 2-cpm background, this equates to 3.7 and 171 gross cpm for 0.1 DAC and 10.0 DAC respectively. The σ_n for each case is calculated as follows:

0.1 DAC

$$\sigma_n = 1.96\sqrt{\frac{3.7}{45} + \frac{2}{10}} = 1.04\ cpm$$

Therefore, the resulting ncpm of 1.7 has an error of ±1.04 cpm. This is converted to a %error by; [(1.04/1.7)100%], or 61.3%

10 DAC

$$\sigma_n = 1.96\sqrt{\frac{171}{1} + \frac{2}{10}} = 25.7\ cpm$$

Therefore, the resulting ncpm of 169 has an error of ±25.7 cpm. This is converted to a %error by; [(25.7/169)100%], or 15.2%.

The %error associated with the ncpm is 61.3% for 0.1 DAC and 15.2% for 10 DAC.

Instrument Efficiency

The initial error associated with instrument efficiency is based on its calibration to a NIST traceable source. Typically, the sources at SRS are ordered with an error less than about ±5%. However, ANSI N323A-1997 [ANSI 1997] allows an accuracy of up to ±10% for NIST traceable sources. As there is no specific requirement that the calibration sources be less than ±5%, the error allowed by ANSI [1977] is used. Using the error allowed by ANSI [1997], the allowable error for the calibration source is ±10%.

Once the instrument is calibrated against the calibration source, a series of counts using a check source are performed with the mean and standard deviation determined. For a counting system that is operating within SRS guidance, the standard deviation should be within 3.0% of the mean. The daily checks allow the system to remain operational as long as the source check does not exceed the range of two standard deviations above or below the mean. This equates to an additional error of ±6%.

The total instrument efficiency error (*eff_E*) is propagated using equation 1.0 as follows:

$$\% \mathit{eff}_E = \sqrt{(10\%)^2 + (6\%)^2} = \pm 11.7\%$$

The %error associated with the instrument efficiency is ±11.7%

Flow Measurement

The air samplers at SRS use rotameters to determine the volume of the sample. Rotameters at SRS are calibrated to within ±15% of the standard used. In addition, the error associated with the TSI velocity meter used to check the rotameter is about ±3% [TSI 1998]. Additional error can be introduced from improper use of the TSI during calibration. For purposes of this work, proper use of the TSI was assumed.

Additional error is introduced when personnel read the rotameter. When reading a meter scale, as in a rotameter, the arbitrary uncertainty is customarily assumed to be one-half of the smallest scale division on the instrument [NRC 1993]. The scale for rotameters used in the air sampler is 0.5 cfm per division. Therefore the potential error is (0.5 cfm/2) or 0.25 cfm. Typical flowrate for an air sampler is 3-5 cfm; therefore the % error is calculated for the average flowrate of 4 cfm. The error associated with reading the rotameter is [(0.25/4)(100%], or ±6.3%.

The total flow measurement error ($\dot{V}_E$) is propagated using equation 1.0 as follows:

$$\% \dot{V}_E = \sqrt{(15\%)^2 + (3\%)^2 + (6.3\%)^2} = \pm 16.3\%$$

The %error associated with the flow measurement is ±16.3%

Collection Efficiency

Normally, the collection efficiency would be considered a constant, however, Regulatory Guide 8.25 [NRC 1992] states "If the efficiency of the collection media (such as filters) for an air sample is less than 95 percent for the material being collected, the sample result should be corrected…". Using this guidance, an error up to -5% could be introduced into the analysis for filter papers with a collection efficiency of ≥95%.

The %error associated with the collection efficiency is -5%.

Sample Time

When using a timing device to measure sample volume, an appropriate value of the percent uncertainty for usual sampling intervals is 1% [NRC 1993].

The %error associated with the sample time is ±1.0%

Self Absorption

Previous work by Higby [1984] showed that the factor used at SRS should be a conservative value thus providing a positive error. However, work by Luetzelschwab et al [2000] indicates that the SA may not be as conservative as previously shown.

In both cases the actual error associated with the SA value is not known with any precision. As such, a ±10% Process Measurement (PM) uncertainty is assumed for SA. The use of the PM uncertainty gives a good starting value for the possible error associated with a particular input.

Once the overall error analysis is performed, it can be determined how much the use of the PM uncertainty SA error contributed to overall error. If it contributes significantly, more research should be performed to derive a more defensible value. If it contributes little and nothing indicates that the value should be higher, nothing more needs to be done.

The %error associated with self-absorption is ±10.0%

Total Error Determination

The total error is calculated using the previously determined %error for each of the input values. Since the ncpm required examples to determine a %error, the total error, using equation 1.0, is calculated for each example.

0.1 DAC

$$\%E_T = \sqrt{(61.3\,(ncpm))^2 + (11.7\,(eff))^2 + \left(16.3\,(\dot{V})\right)^2 + (1\,(t_s))^2 + (5\,(CE))^2 + (10\,(SA))^2} = 65.5\%$$

10.0 DAC

$$\%E_T = \sqrt{(15.2\,(ncpm))^2 + (11.7\,(eff))^2 + \left(16.3\,(\dot{V})\right)^2 + (1\,(t_s))^2 + (5\,(CE))^2 + (10\,(SA))^2} = 27.6\%$$

OPTIMIZATION

The total errors for a representative RAS using the examples in this memorandum were 65.5% and 27.6% for the 0.1 and 10 DAC examples respectively. The largest error was that associated with low level counting where the error associated with counting statistics dominates the total error.

Increasing the background or sample count times can reduce the error associated with counting statistics (reducing background will also work but may not be as practicable). In the 0.1 DAC example, the sample was counted for 45 minutes. Increasing the sample count time would not be practical for this example. However, the background (performed once per day) could be increased to 30 minutes. This would reduce the σ_n to 0.76 cpm, the %error to 44.5%, and the total error to 50.1%. A reduction in total error of >15%.

In the 10.0 DAC example, the sample was only counted for 1 minute. Increasing the sample count time to 2 minutes would reduce the σ_n to 9.3 cpm, the %error to 5.5%, and the total error to 23.6%. A reduction in the total error of 4%. Note that with the two minute count time, the total error in the 10.0 DAC example is now dominated by sources other than the counting statistics. Therefore, increasing the count time beyond 2 minutes would have little effect on the total error.

CONCLUSION

The errors calculated in this memorandum are intended to give an approximation of the expected error for both low level and higher level radioactivity samples associated with RAS. Due to the random nature of the error associated with counting a radioactive air sample, a specific analysis needs to be performed to determine the overall error for any specific air sample. The methodology and assumptions contained within this paper can be used for this type of calculation.

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