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STATISTICAL ASPECTS OF DETERMINING COMPLIANCE
WITH RADIATION STANDARDS

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Abstract

Radiation surveys are an important tool used to monitor the safety of operations at nuclear fuel cycle facilities, as well as determining if contaminated sites require remedial action before license termination or unrestricted release. It is important that radiation surveys are carefully designed to provide the right quantity and quality of useful information for making valid decisions concerning public safety. The validity of survey information is especially important when low-level radiation detection techniques are required such as for environmental radiation monitoring. Thus, statistical aspects of radiation surveys are important in demonstrating compliance with radiation guidelines and for deciding when remedial action or cleanup is required. In this paper, we discuss the statistical aspects of evaluating whether guidelines are, in fact, being exceeded.

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Introduction

United States Federal regulations require environmental monitoring of light-water reactor power stations and other licensed nuclear facilities to ensure compliance with radioactivity release limits (CFR50)(CFR20). Specifically, 10 CFR Part 20 requires that a licensee conduct physical surveys "as may be necessary for him to comply with the regulations in this part." Guidance on environmental sampling techniques to help meet these regulations is found in the U.S. Nuclear Regulatory Commission (NRC) Regulatory Guides and in procedures developed by the U.S. Department of Energy (DOE) Environmental Measurements Laboratory (RG4.5)(RG4.6)(RG4.8)(Ha77). Further guidance on allowable surface contamination levels for unrestricted public release of property from a licensed nuclear facility is found in American National Standard Institute (ANSI) Standard N13.12 and NRC Regulatory Guide 1.86. (AN13.12)(RG1.86).

These radioactivity contamination guidelines and regulations have evolved as a means of protecting public and occupational health and safety. Such attempts at legislating "as low as reasonably achievable" (ALARA) conditions are successful only when members of the nuclear industry design their radiation monitoring programs to ensure compliance. In situations where it is difficult to use conventional statistical techniques or standard radiation detection methods, demonstrating compliance to these standards can be a major problem. For either detecting low energy beta-gamma emitters or measuring low levels of contamination such as those encountered in environmental radiation monitoring, difficulties in demonstrating compliance can create a callous attitude in

radiation survey programs which may lead to situations that directly conflict with the philosophy of ALARA.

Literature on industrial quality control presents a large number and wide variety of methods that could be useful for radiation sampling programs. However, the low-level special sampling aspects of proving compliance with guidelines are not always analogous to the industrial quality control problem of detecting a faulty item in an inventory. Thus, in the first part of this paper we discuss some of the difficulties of using basic quality control sampling methods for these cases.

Radiation survey techniques used in low-energy or environmental monitoring programs should be selected to give high quality information. The choice of instrumentation, counting time, and background counting conditions are important to determine the best survey technique for conducting a radiation survey. In the second part of the paper we present a general discussion of radiation detection considerations for low energy or low-activity samples.

Statistical Sampling Methodologies

All statistically rigorous sampling plans require that the risk of making two types of mistakes be specified in advance. In quality control terminology, the mistake of concluding that manufactured items are acceptable when in fact they are defective is called consumers risk. The mistake of concluding such items are defective when in fact they are good is called producers risk. The radiation monitoring analog of consumers risk is to allow a facility to continue

operating when in fact it is releasing amounts of radioactivity that exceed standards or regulations. The analog of producers risk is to shut down a facility when there is no true reason to do so. Because of the statistical nature of all measurements, especially radioisotope counting procedures, it is impossible to reduce either risk to zero.

The science of quality control has developed an extremely useful method of plotting or visualizing these risks and relating them to the true level of manufactured item quality, or analogous innocuous levels of radioactive contamination. This method is called the "operating characteristics curve," or more often the "OC curve" (We77). The shape of this curve is controlled by the levels of the two kinds of risk that are arbitrarily specified, the variance of the measuring procedures, the variance of the process being measured, the sample sizes, and the range of acceptable item quality levels. Typically in industrial or manufacturing situations, the producers and consumers risks are specified to be around 5 to 10 percent.

Once an OC curve has been specified, a number of sampling strategies can be used to determine sample size and decision rules. These strategies vary in how the samples are to be collected and in how the results are analyzed. The standard strategies include the use of control charts, cumulative sums and single, double or sequential sampling methods (We77). The rather difficult Bayesian and Decision Theory approaches are more complicated strategies (We77), but they have increased efficiency in the sense that smaller numbers of replicate samples can be used. For environmental sampling, the sequential methods

are usually impractical because each sample has to be analyzed before the next one is collected; such repeated sampling is much less economic than collecting many replicate samples at one time. The Bayesian and Decision Theory approaches require making many assumptions about the statistical structure of the data as well as specifying the cost of making the wrong decision. For most environmental radiation surveys it is not economical, in time or money, to verify the assumptions and find the needed cost. Thus, the only practical statistical methods for controlling errors seems to be the very standard control chart strategies.

The number of replicate samples needed for radionuclide work is a function of the counting error, the relative accuracy desired, and the assigned risk of making the two kinds of errors discussed above. In radionuclide counting it is well known that the counting error can be made arbitrarily small by using sufficiently long counting times. In modern instruments this is accomplished by measuring the time needed to reach a predetermined total number of counts. Thus, it is always possible to adjust the counting part of the analysis so only that one replicate sample can be used. Statistically this is not a very useful procedure; it assumes the only analytical and sampling error is that of the radioactive decay process. In environmental monitoring it is necessary to also consider the errors introduced by sample preparation, and the variability between collected samples. These errors cannot be controlled and are usually significant contributors to the total error. Sampling error is measured as the standard deviation, after counting and analytical error are subtracted out, among several

samples collected at one sampling site. Analytical error is measured as the standard deviation of several aliquots of one sample, after counting error is subtracted out.

The simplest and most common way of determining the necessary sample size for a specific sampling and analytical error is to use the "sample size for estimation" formula (Di69):

$$N = \left(\frac{ZS}{d}\right)^2 \quad (1)$$

where N is the number of samples, Z is a constant taken from a table of the normal distribution for the desired confidence level (typically 95% confidence is used), S is an estimated total standard deviation (total error for the number of samples, counting, and analytical error for the number of aliquots), and d is the desired accuracy expressed in the units of measurement. The difficulty with this approach is that S, the total standard deviation, must be estimated before an experiment to estimate S can be conducted. It seems that the answer to the problem must be known before S can be estimated. The estimate of S is usually a guess based upon similar situations in the past; thus, the experiment gives a refined estimate. If the original guess is bad, then the results of the first experiment are used to find the sample size for a second experiment. In other words, nothing reasonable can come immediately from a situation of complete ignorance. A somewhat more useful form of this formula makes use of the coefficient of variation ($CV = \text{standard deviation} \div \text{mean value}$) and the relative accuracy ($A = d \div \text{mean value}$). In radionuclide work it

is customary to express the error as a percent of the mean. When these percentages are substituted into the formula for N, the mean values cancel out giving (Eb78):

$$N = \left(\frac{Z \text{ CV}}{A} \right)^2. \quad (2)$$

The above formulas, listed in equations 1 and 2, consider only one type of error. If both the consumers and the producers risks are to be controlled, the Z in these equations is replaced by $(Z_1 + Z_2)$ which are the normal deviates tabulated for the desired risk of making the two types of errors.

The counting time required to achieve a given counting error can be derived from the basic equations for a Poisson distribution. For a Poisson distribution, the mean value equals the variance ($m = \sigma^2 \cong n^2$), and if n is the number of counts recorded by an instrument over time t the counting rate is $r = n/t$. The count rate $\pm$ its standard deviation is:

$$r \pm \sigma_r = \frac{n}{t} \pm \frac{n^{1/2}}{t} = r \pm \left(\frac{r}{t} \right)^{1/2}. \quad (3)$$

In terms of percentage error:

$$r \pm \frac{100}{(rt)^{1/2}} \% = \frac{100}{n^{1/2}} \%. \quad (4)$$

Thus, the percentage error of a counting measurement is determined by the total number of counts accumulated (Pr64).

When several independent quantities are combined by addition or subtraction, the standard deviation is:

$$\sigma_s = (\sigma_1^2 + \sigma_2^2 + \dots + \sigma_n^2)^{\frac{1}{2}} \quad (5)$$

Thus, for a background counting rate of $r_b \pm \sigma_b$ and a total counting rate (source plus background) of $r_T \pm \sigma_T$, the source counting rate is:

$$r_s \pm \sigma_s = (r_T - r_b) \pm (\sigma_b^2 + \sigma_T^2)^{\frac{1}{2}} \quad (6)$$

By substitution from Equation 3:

$$\sigma_s = \left(\frac{r_b}{t_b} + \frac{r_T}{t_T} \right)^{\frac{1}{2}} \quad (7)$$

To determine the optimum use of counting time, Equation 7 is differentiated with respect to time, solved for minimum error ($\frac{d\sigma_s}{dt} = 0$) assuming a constant time ($t_b + t_t = \text{a constant}$) (Pr64). The result is:

$$\frac{t_b}{t_T} = \left(\frac{r_b}{r_T} \right)^{\frac{1}{2}} \quad (8)$$

The fractional accuracy, d , is found by:

$$d = \frac{\sigma_s}{r_s} = \frac{\sigma_s}{(r_T - r_b)} = \frac{(r_b/t_b + r_T/t_T)^{\frac{1}{2}}}{(r_T - r_b)} \quad (9)$$

Solving this equation for the sample counting time, t_T , gives:

$$t_T = \frac{\left[1 + \left(\frac{r_b}{r_T} \right)^{\frac{1}{2}} \right]^2}{r_T d^2 \left(1 - \frac{r_b}{r_T} \right)} \quad (10)$$

Thus, the required counting time associated with various levels of contamination for a given accuracy, d , can be estimated. As an example,

Equation 10 is solved for an accuracy of 5% for a background count rate of 30 counts per minute, and an assumed $\beta^- \cdot \alpha$ detector efficiency of 3%. The results of this example calculation are shown in Table 1 for various levels of surface contamination on smears over an area of $3 \times 10^{-2} \text{ m}^2$. This example shows that extremely long counting times are required to detect low-levels of radioactive contamination. A more detailed discussion of the detection of low levels of radioactivity is found in a paper by Donn and Wolke (Do77).

Instrumentation Considerations at Long Counting Times

Several important factors influence the ability to detect the levels of radiation required to prove compliance with current radiation safety standards. These factors include: the level of background radiation, the specific activity of the radioactive source, the types and energies of the radiation emitted from the source, the source-detector geometry, the instrument time constant, the speed at which the detector is moved across the source, and the detection efficiency of the instrument used. Recent studies have addressed the problem of measuring radioactive contamination with common field instrumentation (Bu71)(So75)(Il77). In general these studies have reported that special problems arise when low-energy emitting radionuclides are present in the mixtures. When the detection efficiency is small because of these low-energies, long counting times are required to accurately determine the level of contamination (as shown by the example problem results in Table 1 with an assumed detection efficiency of 3%). In the field it is difficult to maintain a constant source-detector

Table 1. Calculated Counting Time Requirements for Surface Contamination Smears

Surface Contamination Level (dpm/10 ⁻⁴ m ²)	Activity Per Smear (dpm/3x10 ⁻² m ²)	Total $\beta^- \cdot \alpha$ Count Rate (Counts/Minute) ^(a)	Counting Time (5% Accuracy)	
			Sample	Background
220	66,000	2,010	1.4 sec	1.7 sec ^(b)
22	6,600	228	3.2 min	1.1 min
2.2	660	50	93 min	72 min
0.22	66	32	4.9 days	4.8 days

(a) The total count rate is found for a $\beta^- \cdot \alpha$ detector with an assumed detection efficiency of 3%, plus an assumed background count rate of 30 counts/minute.

(b) For sample activities much greater than background, error in the background count may be neglected.

geometry that matches the time constant of the instrument used when long counting times are required. The detector response can be improved by lowering the background count rate, but this usually means that a laboratory with a shielded counting volume is required. Thus, the cost of adequately proving compliance with current regulations for low-energy radionuclides may be quite high.

In situations where it is difficult to use common field instruments for radiation surveys, workers may develop a callous attitude towards the importance of these surveys. This attitude could reflect in poor technique and result in working conditions that conflict with the philosophy of ALARA. Since the intent of ALARA is to assure public and occupational safety, care must be taken when radiation survey programs are established. Periodic review of survey techniques and instrumentation may help prevent deterioration of the workers attitude.

Statistical Problems at Long Counting Times

Whenever the counting time becomes a substantial fraction of the half life of any isotope in a sample, two new problems arise: the counting rate may decrease significantly during the time of counting, and there may be significant daughter product ingrowth during this same time.

The rate of decay is a measure of the slope of the activity level versus time curve. It is estimated by dividing the total counts by the duration of counting. If there is a significant decrease in the activity level over the duration of the count, the counting rate will also change over the duration of the counting. Thus, more counts will accumulate in the first part of counting than in the last part.

The calculated rate of decay is an average or integration over the duration of the counting period. For purposes of extrapolating back to the time of sample collection or to some sample extraction step, we need to assign a specific time at which the calculated rate of decay occurred. For long counting times relative to a half life, neither the time that the count was started or was ended is typical of the calculated rate. Something in between should be used. The time at which half the counts occurred before and half after is a median time, and is the time at which the calculated decay rate was the instantaneous true rate of decay. Applying some calculus to the algebraic formula for exponential rate of decay results in the following formula:

$$t_m = \frac{1}{R} \left[\ln 2 - \ln \left(e^{-Rt_E} + e^{-Rt_B} \right) \right] \quad (11)$$

where: t_m = median time

t_B = time of beginning of counting

t_E = time of ending of counting

R = rate of isotope decay = $\ln 2 \div \text{half-life}$

$\ln$ = natural logarithm.

The low concentrations of isotopes such as Th-228 and Ra-226 found in many environmental samples require counting times up to 20 hours to attain reasonable statistical accuracy. These isotopes are assayed by separating out the Ra-224 and Rn-222 daughters and measuring the daughter products. For these or any radioactive series, the counts follow a Poisson law only if the counting interval is much shorter than the mean half life of all nuclides being counted. If this condition

does not hold, the standard deviation in the counts is not accurately estimated by the square root of the counts.

A good procedure for handling such situations has been developed by Lucas and Woodward (Lu63). In the following paragraphs we will review the logic of their method. In order to use the method, it is necessary to use tables given in their Argonne report.

When counting, particularly alpha activity, of long decay chains in which the mean life of some of the nuclides is very short compared to the counting interval, and in which the solid angle subtended by the detector is large, there is very high probability of observing counts not only from the parent nuclide but also from the daughters. If this observed count were used to estimate the activity of the parent nuclide, the estimate and its standard deviation would be too high. Lucas and Woodward calculate the ratio of observed counts to those from the parent atoms. Dividing the observed counts by this ratio yields the activity of the parent nuclide.

The value of this ratio is obtained by mathematically modeling the decay chain and statistically modeling the probability that a decay is counted. The value of this ratio varies from isotope series to isotope series, with duration of the counting interval, with solid angle and efficiency of the detector, and with time after sample separation or purification. Lucas and Woodward give tables of some of these variable conditions for radium-224 and radon-222. They provide the mathematical formulas for calculating this ratio for any situation.

Summary

Federal regulations and industrial guidance on allowable radioactive contamination levels have evolved as a means of protecting public and occupational health and safety. The low levels of radioactivity defined by current regulations can be difficult to detect for environmental samples. In designing a radiation survey program, consideration of statistical and instrumentation difficulties must be a priority. Both consumers risk and producers risk must be considered in radiation survey programs to ensure that facilities are safely operated. After reviewing common quality control techniques for plotting these risks, it is concluded that most sequential sampling methods are impractical since single samples must be analyzed before further numerous replicate samples are collected. Also, more complicated approaches, such as Bayesian or Decision Theory analyses, require extensive assumptions that are costly and difficult to verify. Standard Control chart strategies appear to be the only practical statistical methods for controlling errors. Consideration must also be given to the number of replicate samples that need to be taken to achieve a specific sampling and analytical error.

When long counting times are required, as predicted by equations based on the Poisson distribution for low-activity samples, special instrumentation problems arise. When long counting times are required, adequate instrumentation is chosen based on factors such as: the background count rate, the total counting time required, the source-detector geometry, and the types and energies of the radiation emitted from the source. Also, when the counting time of a sample becomes a

substantial fraction of the half-life of any radionuclide in the sample, special corrections must be made to determine the sample activity. Corrections must be made for the influence of radioactive daughter products that may interfere during long counts.

Attempts at legislating "as low as reasonably achievable" conditions are only successful when radiation surveys in and around nuclear facilities are designed to ensure compliance.

REFERENCES

- AN13.12 Draft American National Standard N13.12, 1977, Control of Radioactive Surface Contamination on Materials, Equipment, and Facilities to be Released for Uncontrolled Use, ANSI N13.12, American National Standards Institute, Inc. (New York).
- Bu71 Bush, D. and Hundal, R. C., 1971, "Difficulties in Measuring the Maximum Permissible Levels of Surface Contamination for Radionuclides that Decay Primarily by Electron Capture", Health Physics, 21, 651.
- CFR20 "Standards for Protection Against Radiation", 1979, U.S. Code of Federal Regulations, Title 10, Part 20, Superintendent of Documents, GPO (Washington, D.C.).
- CFR50 "Licensing of Production and Utilization Facilities", 1979, U. S. Code of Federal Regulations, Title 10, Part 50, Superintendent of Documents, GPO (Washington, D.C.).
- Di69 Dixon, W. J. and Massey, F. J., Jr., 1969, Introduction to Statistical Analysis, Third Edition, (New York: McGraw-Hill) 80.
- Do77 Donn, J. J. and Wolke, R. L., "The Statistical Interpretation of Counting Data from Measurements of Low-Level Radioactivity", Health Physics, 32, 1.
- Eb78 Eberhardt, L. L., 1968, "Appraising Variability in Population Studies", J. Wildlife Management 42, 207-238.
- Ha77 Harley, J. H., Ed., 1977, HASL Procedures Manual, HASL-300, Supplement 5, Health and Safety Laboratory (New York, NY).

- I177 Iles, W. J., Burgess, P. H., and White, D. F., 1977, Measurement of Activity of Surfaces Contaminated by Beta-Emitting Nuclides and ^{55}Fe , National Radiological Protection Board, NRPB-R60 (Harwell, England).
- Lu63 Lucas, H. F., Jr., and Woodward, D. A., 1963, Argonne National Laboratory, Radiological Physics Division, Semiannual Report, ANL-6646.
- Pr64 Price, W. J., 1964, Nuclear Radiation Detection - Second Edition (New York: McGraw Hill), 59-62.
- RG4.5 Regulatory Guide 4.5, 1974, Measurements of Radionuclides in the Environment Sampling and Analysis of Plutonium in Soils, U.S. Atomic Energy Commission, Directorate of Regulatory Standards.
- RG4.6 Regulatory Guide 4.6, 1974, Measurements of Radionuclides in the Environment Strontium-89 and Strontium-90 Analysis, U. S. Atomic Energy Commission, Directorate of Regulatory Standards.
- RG4.8 Regulatory Guide 4.8, 1975, Environmental Technical Specifications for Nuclear Power Plants, U. S. Nuclear Regulatory Commission, Directorate of Regulatory Standards.
- RG1.86 Regulatory Guide 1.86, 1974, Termination of Operating Licenses for Nuclear Reactors, U.S. Atomic Energy Commission, Directorate of Regulatory Standards.
- So75 Sommers, J. F., 1975, "Sensitivity of G-M and Ion Chamber Beta-Gamma Survey Instruments", Health Physics, 28, 755.
- We77 Wetherill, G. B., 1977, Sampling Inspection and Quality Control (New York: John Wiley & Sons).