Appendix A

Statistical Treatment of Assay Data

This appendix provides a brief discussion of the statistical treatment of nondestructive assay data. It contains several useful statistical formulas and procedures for estimating assay errors. The discussion considers random errors (assay precision) only. There is no consideration of the often serious problem of systematic errors (assay bias). For a more thorough discussion of assay precision and bias, please refer to textbooks on statistics.

A.1 GENERAL DEFINITIONS

Assume that some physical quantity x is measured N times, with the results x_1, x_2, x_3, ..., x_N. For example, x could be the plutonium mass of a sample measured with a neutron well counter. The best estimate of the true value of x is the average, or mean value,

$$\bar{x} = \sum_{i=1}^{N} x_i / N \tag{A-1}$$

In general, each individual measurement x_i deviates from the mean. A common indicator of the magnitude of this deviation is the standard deviation

$$\sigma = \sqrt{\frac{\sum_{i=1}^{N} (x_i - \bar{x})^2}{N - 1}} \quad (N > 1) \tag{A-2}$$

The estimated standard deviation is often quoted as the relative standard deviation (RSD), which is given by

$$\sigma_r(\%) = (\sigma / \bar{x}) 100 \tag{A-3}$$

It is usually assumed that the measurements are distributed about the mean according to a Gaussian (or normal) distribution. An example of the Gaussian distribution is shown in Figure A.1, which is a histogram of 500 measurements with a Gaussian shape superimposed. The mean value of the measurements is 107.3, and the standard deviation σ is 2.43. The abscissa is in units of σ. For a Gaussian distribution, the full width at half maximum height (FWHM) is 2.354σ. One can also estimate the percentage of the measurements that should lie within a specified interval about the mean. Table A-1 summarizes the estimated percentages in units of σ. The distribution of measurements shown in Figure A.1 is very close to these estimates.

Table A-1. Percentage of measurements expected to lie within $\pm w\sigma$ of the mean of a Gaussian distribution

Width of Region, $\bar{x} \pm w\sigma$	Estimated Percentage of Measurements in Region
$\pm 0.6745\sigma$	50.00%
$\pm 1.0000\sigma$	68.27%
$\pm 2.0000\sigma$	95.45%
$\pm 3.0000\sigma$	99.73%

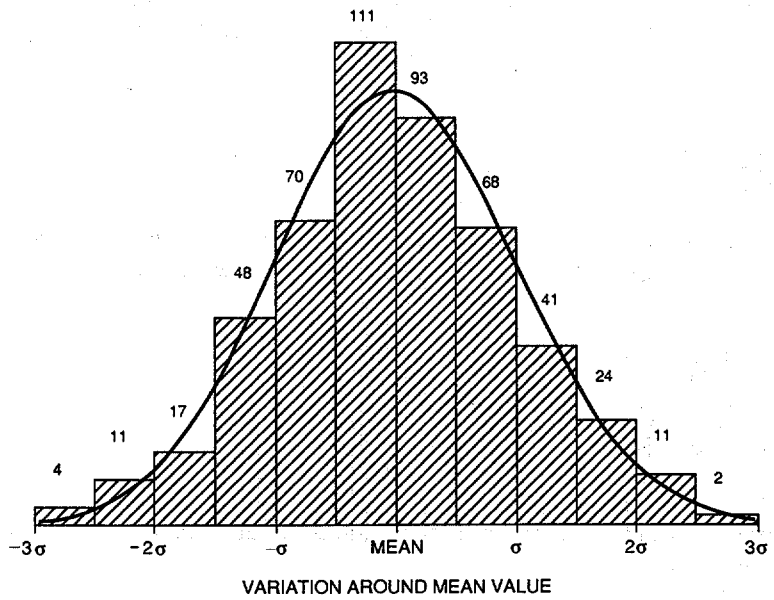

Fig. A-1. *A histogram of 500 measurements distributed about a mean. The solid line is a superimposed Gaussian shape.*

The mean value $\bar{x}$ calculated from Equation A-1 is subject to some measurement uncertainty. The estimated standard deviation of the mean that is determined from N measurements is

$$\sigma_{\bar{x}} = \sigma/\sqrt{N} \quad . \tag{A-4}$$

This equation indicates that the mean is determined more precisely as the number of measurements N increases. From Table A-1, there is a 68% probability that the true mean lies within the range $\bar{x} \pm \sigma/\sqrt{N}$ and a 95% probability that the true mean lies within the range $\bar{x} \pm 2\sigma/\sqrt{N}$.

The standard deviation σ calculated from Equation A-2 is also subject to measurement uncertainty. The standard deviation of the standard deviation follows a chi-square distribution. An approximate equation for the RSD of σ that is correct to about 10% for N greater than 3 is

$$\text{RSD of } \sigma \approx 1/\sqrt{2(N-1)} \, . \tag{A-5}$$

Table A-2 provides a more accurate compilation of the probability that the standard deviation lies within a given interval. (From Table A-1 it can be seen that the interval in Table A-2, 90% probability, has a width of almost 2σ.) Equation A-5 and Table A-2 show that the standard deviation, like the mean, will be determined more precisely as the number of measurements increases, but that there is a large variation in the computed standard deviation even for 20 or 30 repeated measurements.

Table A-2. Standard deviation of the standard deviation for a series of repeated measurements. For example, for 10 measurements, there is a 90% probability that the true standard deviation lies in the interval 0.74σ to 1.59σ, where σ is the standard deviation estimated from Equation A-2

Number of Measurements	Lower Limit of Interval 5% Probability	Upper Limit of Interval 95% Probability
2	0.58	4.41
3	0.62	2.92
4	0.65	2.37
5	0.67	2.09
7	0.71	1.80
10	0.74	1.59
15	0.77	1.44
20	0.80	1.36
25	0.81	1.31
30	0.83	1.27

A.2 PROPAGATION OF ERRORS

Often the final answer, such as grams plutonium, involves several different measurements with different uncertainties. For example, suppose that plutonium mass m = C(P – kB), where C = calibration constant, P = counts in peak window, k = a constant, and B = counts in background window. The variables C, P, and B may all have different uncertainties, which must be combined, or propagated, to arrive at the final error in the mass.

There are several common formulas that can handle most simple combinations of errors. Let $x \pm \sigma_x$ and $y \pm \sigma_y$ be two independent variables, and let k be a constant with no uncertainty.

$$\text{If } z = x + y \text{ or } x - y, \quad \sigma_z = \sqrt{\sigma_x^2 + \sigma_y^2} \; . \tag{A-6}$$

$$\text{If } z = x/y \text{ or } xy, \quad \frac{\sigma_z}{z} = \sqrt{\left(\frac{\sigma_x}{x}\right)^2 + \left(\frac{\sigma_y}{y}\right)^2} \; . \tag{A-7}$$

$$\text{If } z = kx, \quad \sigma_z = k\sigma_x \; . \tag{A-8}$$

For example, for m = C(P – kB),

$$\frac{\sigma_m}{m} = \sqrt{\left(\frac{\sigma_C}{C}\right)^2 + \frac{\sigma_P^2 + k^2\sigma_B^2}{(P - kB)^2}} \; . \tag{A-9}$$

Other formulas for error propagation can be derived by differentiating the equation z = f(x,y) and squaring the result:

$$(dz)^2 = \left(\frac{\partial z}{\partial x}\right)^2 (dx)^2 + \left(\frac{\partial z}{\partial y}\right)^2 (dy)^2 + 2\left(\frac{\partial z}{\partial x}\frac{\partial z}{\partial y}\right)(dx\ dy) \; . \tag{A-10}$$

The cross term contains the product (dx dy). If x and y are independent variables, then dx and dy are uncorrelated. If a series of measurements are made to determine z, then the measurement uncertainties dx and dy fluctuate randomly between positive and negative values, and the cross term (dx dy) has an average value close to 0. Also, the average of squared differentials like $(dx)^2$ is the square of the standard deviation, σ_x^2. Then the square root of Equation A-10 becomes

$$\sigma_z = \left[\left(\frac{\partial z}{\partial x}\right)^2 (\sigma_x)^2 + \left(\frac{\partial z}{\partial y}\right)^2 (\sigma_y)^2\right]^{1/2} \; . \tag{A-11}$$

Equations A-6, A-7, and A-9 can be derived from Equation A-11, as can any other equation needed for more complex error propagation.

A.3 Nuclear Counting Statistics

For measurements involving nuclear particle counting, all of the above information can be applied. In addition, in a nuclear counting measurement, the radioactive decays or other randomly-spaced events usually follow a Poisson distribution, for which the standard deviation σ_x of a single measurement can be estimated by

$$\sigma_x \approx \sqrt{x} \tag{A-12}$$

where x is the actual number of counts. Note that Equation A-12 applies to counts and not to count rate. If a count rate is measured for a time t, yielding a single measurement of x, there is a 68% probability that the actual rate is included in the interval $(x \pm \sqrt{x})/t$.

Consider the example of $m = C(P - kB)$. Assume that k=1 and that σ_C=0.

$\sigma_P \approx \sqrt{P}$, $\sigma_B \approx \sqrt{B}$, and $\sigma_m \approx C\sqrt{P+B}$.

The RSD (in percent) is

$$\sigma_r(\%) = \frac{\sigma_m}{m} \approx 100\frac{\sqrt{P+B}}{P-B}. \tag{A-13}$$

If N measurements are made on the same sample, the RSD of the distribution σ_r can be calculated from Equation A-2 (with m_i replacing x_i) and Equation A-3, or it can be estimated from

$$\sigma_r(\%) \approx 100\frac{\sqrt{\bar{P}+\bar{B}}}{\bar{P}-\bar{B}} \quad . \tag{A-14}$$

The two ways of computing σ_r should yield similar results if the number of repeat measurements, N, is large. If the results are not similar, the counting equipment may be malfunctioning.

Note that all of the discussion in this appendix pertains to the precision or repeatability of measurements. This analysis gives no information regarding the accuracy of a measurement (how well the measurement determines the correct amount of material).

Appendix B

Radiation Safety

The passive nondestructive assay (NDA) techniques described in this book rely on the natural radiation emitted by nuclear material. The assayist should be aware of the amount and type of radiation being emitted by the sample to ensure that the measurement does not pose a safety hazard. This appendix provides some background information on radiation safety and gives some examples of typical sample dose rates.

The radiation emitted by plutonium, uranium, thorium, and reactor fission products consists of alpha particles, beta particles, x rays, gamma rays, and neutrons. Because the alpha particles have a very short range (3-4 cm in air), they do not present a health hazard unless the active material is inhaled or ingested. When monitoring for alpha-particle contamination, the radiation meter must be held very close to the surface. Alpha-particle radiation is usually measured with an ionization chamber that has a very thin metal foil window. Beta particles have a range of several millimeters in most materials, and x rays and gamma rays have ranges of several centimeters or more. A typical beta-gamma meter has a Geiger tube or thin scintillator and a sliding metal window that is opened for measuring beta particles and closed for measuring x rays or gamma rays. Neutron radiation is more penetrating and more hazardous than any of the other radiations and is usually detected with a ^3He or BF_3 detector surrounded with a 20-cm-diameter sphere of polyethylene (a Bonner sphere or "cow").

Radioactive material is usually characterized by its activity or disintegration rate, as measured in curies. One *curie* (Ci) is 3.7×10^{10} disintegrations per second. The amount of energy deposited, the absorbed dose, is given in units of rads. One *rad* is a quantity of radiation that leads to the absorption of 100 ergs (624 200 MeV) per gram of irradiated material. The biological damage produced by a dose of 1 rad varies with the rate of energy loss in tissue. To determine the equivalent dose from different kinds of radiation, one uses the unit *rem* defined as

$$\text{rem(equivalent dose)} = \text{QF} \times \text{rad(absorbed dose)}.$$

Values for the quality factor QF are given in Table B-1. The International Commission on Radiation Protection has recommended that the quality factor for fast neutrons be increased to 20, but as of January 1989 the U.S. Department of Energy recommends that, based on the available data, the quality factor remain at 10. A new international unit of equivalent dose, the *sievert*, is equal to 100 rem.

Table B-1. Quality factor QF for the equivalent dose of different types of radiation

QF =	1	beta, x, gamma radiation
	2.3	thermal neutrons
	5	protons
	10	alpha particles
	10	fast neutrons
	20	massive charged particles like fission fragments

There are several approximate relationships that can be used to convert the strength of gamma-ray and neutron sources into dose rates. For a gamma-ray source of energy E (in MeV) and strength C (in curies),

rem/h at 30 cm $\approx$ 6CE.

For a fast-neutron source, the exposure rate is

$\sim$1 millirem per hour (mrem/h) at 1 m per 10^6 n/s

For a thermal-neutron source, the exposure rate is

$\sim$0.1 mrem/h at 1 m per 10^6 n/s .

Examples of typical dose rates encountered in passive NDA assay are given in Table B-2. The plutonium dose rate may be much higher if the americium content is more than 0.1%.

Table B-2. Some typical dose rates encountered in passive NDA

Radiation Source	Source Strength	Dose Rate at 10 cm (mrem/h)		Dose Rate at 1 m (mrem/h)	
		Neutron	Gamma	Neutron	Gamma
1 μ g ^{252}Cf	2.3 $\times$ 10^6 n/s	230	14	2.3	0.14
100 μCi ^{137}Cs	3.1 $\times$ 10^6 γ/s	0	3.0	0	0.03
PuO_2(6% ^{240}Pu)	1 kg	$\sim$10	$\sim$100	$\sim$0.1	$\sim$1
UO_2 (93% ^{235}U)	1 kg	$\sim$0	1.2	$\sim$0	0.01
Natural bkg	environment	0.01-0.02 (100-200 memr/yr)			

The biological effects of radiation are summarized in Table B-3 for acute (2 hours or less) and chronic (long term) exposures to the whole body. Based on these effects, maximum allowable radiation doses have been established by the International Commission on Radiation Protection. These recommendations are summarized in Table B-4 and may be compared to the natural background radiation level of 0.1 to 0.2 rem/yr. The maximum allowed doses are far below those that would show acute biological effects. Furthermore, in most facilities, worker exposure is held well below the allowed maximum.

The International Commission on Radiation Protection also recommends that the radiation dose should be kept as low as practical or "as low as reasonably achievable (ALARA)." The NDA operator can limit radiation dose from a source in three ways: minimize the exposure time, maximize the distance to the source, and shield the

Table B-3. Biological effects of radiation on the whole body

Dose	Probable Effect
Acute dose below 25 rem	No noticeable effect
Acute dose of 25-75 rem	Blood changes detectable in lab tests
Acute dose above 100 rem	Physical symptoms such as nausea, hair loss
Acute dose of 350 rem	50% fatality rate in 1 month
Acute dose of 600 rem	95% fatality rate
Chronic low-level dose	1 death per 7000 man-rem/yr
Chronic low-level dose	Less than 1% increase in genetic disorders per million man-rem/yr

Table B-4. Maximum allowable radiation doses above natural background

Person	Maximum Dose
Radiation worker	3 rem in 3 months (6 mrem/h continuous in 40-h week) 5 rem in 12 months (2.5 mrem/h continuous in 40-h week)
Pregnant worker	0.5 rem to fetus during pregnancy
General population	0.5 rem in 12 months

source. The operator can measure the dose rate of the source with a health physics instrument or estimate the dose rate by calculation. Unless the dose rate is completely negligible, the operator should minimize the amount of time spent near the source. Because the radiation dose from most sources decreases as the square of the distance, the source should be kept as far away as practical and handled as little as possible. If large sources must be used, then radiation shielding is necessary. Information on gamma-ray attenuation by dense materials is given in Chapter 2, and information on neutron shielding is given in Chapter 12, Section 12.6.

Appendix C

Criticality Safety

The nondestructive assay (NDA) of fissile material often involves placing the sample into a highly reflecting geometry or placing it close to other samples to be assayed. Both of these actions can potentially lead to a criticality accident and fatal radiation exposure. If the proper combination of fissile material, moderators, and reflectors is present, a self-sustaining chain reaction can occur. The NDA user is responsible for the safety of himself and others and should have an awareness of criticality safety. This appendix provides a brief introduction to this subject. Additional information is available in the references listed below. In all situations, the NDA user must consult the Criticality Safety Officer in the facility where the user is working and must follow facility guidelines for handling and storing fissionable material.

Criticality results when the neutron fission process achieves a self-sustaining chain reaction. If the production of neutrons exceeds the loss of neutrons by capture or leakage, the system is said to be supercritical. Criticality depends not only on the quantity of fissile material present (such as ^{235}U or ^{239}Pu), but also on the size and shape of the container, on the nature of any neutron-moderating material present in the container, and on the presence of any adjacent material (including human bodies) that might reflect neutrons back into the container.

The minimum critical masses of some fissionable materials are given in Table C-1. The minimum critical masses occur for spherical geometries, and these masses are lower if the sphere is surrounded by materials that reflect and moderate neutrons. For example, a critical sphere of uranium metal at normal density with an enrichment of 93% ^{235}U has a diameter of about 17.5 cm and a mass of about 49 kg. If the sphere is immersed in water, some of the neutrons are reflected back into the sphere, and the critical diameter drops to about 13 cm, with a corresponding uranium mass of about 22 kg. If sufficient water is also mixed homogeneously with the uranium, the critical diameter increases to 31 cm, but the critical mass of ^{235}U is only 800 g. This last case represents the minimum critical mass of ^{235}U that could be encountered in normal facility processing operations. Table C-1 lists minimum critical masses for three systems: pure metal, pure oxide, and a homogeneous metal-water solution, with the critical mass of each system given bare (no reflectors or moderators) and fully water-reflected (the system is surrounded by an unlimited quantity of water).

Table C-1. Minimum critical masses of some fissionable materials in spherical geometry, bare and fully water-reflected (FWR)

Fissionable Material	Metal (kg)		Oxide (kg)		Solution (g)	
	Bare	FWR	Bare	FWR	Bare	FWR
^{239}Pu(19.7 g/cm^3)[a]	10	5			1000	510
^{239}Pu(14.9 g/cm^3)[a]	16	8	21	14		
^{238}Pu			~30			
^{242}Pu	~80					
^{235}U[b]	49	22	90	43	1600	760
^{233}U	15	7	34	15	1000	500

[a] ^{239}Pu is assumed to be in the form of low-burnup plutonium with approximately 6% ^{240}Pu and 94% ^{239}Pu.
[b] ^{235}U is assumed to be in the form of highly-enriched uranium with approximately 93% ^{235}U and 7% ^{238}U.

Nondestructive assay often places a sample into a highly reflecting geometry for measurement. In particular, passive neutron assay often places the sample into a well surrounded by a thick polyethylene moderator. Some detector wells are lined with cadmium, a neutron poison, but this is not always the case. Although the moderator is not as well coupled to the sample as the fully water-reflected geometry used in Table C-1, it does lead to a measurable increase in neutron reflection and multiplication. The sample itself will usually contain much less than the minimum critical mass of fissionable material, but the NDA operator must be certain that the sample cannot inadvertently contain sufficient material to become critical when placed in the well counter. This can be a difficult problem, particularly for large containers of scrap and waste for which there is no reliable information on the amount of fissionable material, its enrichment, and the matrix in which it is embedded. For small containers of dense material, the operator must also consider the possibility of accidentally placing two containers in the counter.

Another area of concern for the NDA operator is sample storage and transport. It is customary to store many samples in a single vault or safe and to transport them to the NDA instrument in containers that may hold several samples at once. The operator must consider the possibility that, although each individual sample may be critically safe, the storage area or transport container may constitute a stacked array that is not critically safe. Flooding of the array is particularly dangerous, because a flooded array can approach the geometry of a metal-water mixture and, like a reactor fuel assembly, can be much more critical when it is flooded than when it is dry.

The most conservative approach is to rely only on the known gross weight and volume of the sample and assume that the sample-instrument combination constitutes a fully water-reflected geometry. The operator can establish a weight limit for the sample, its transport container, and its storage area that is so low that the given volume could not contain a critical combination of fissionable material and optimum moderator.

If the sample containers are too heavy to meet this conservative limit, there are several other possible ways to arrive at critically safe operating limits. Multiplication measurements may be made inside the assay system (Ref. 1) or neutron transport calculations (such as those described in Chapter 12, Section 12.7) may be carried out using properly validated computational methods (Ref. 2). Many calculations already exist in Refs. 1–7, and some may be applicable to the problem at hand. Another option is administrative control of sample geometry, matrix, or other parameters. If all else fails, it may be necessary to repackage the samples into smaller containers for which critically safe limits can be established.

Regardless of how critically safe limits and operating procedures are established, they must be determined in cooperation with the facility Criticality Safety Officer. This person is an expert because of his experience and training, and the criticality safety of all operations that involve the handling, storage, and measurement of fissionable material are his responsibility as well as the responsibility of the NDA operator.

Considerable information is available on the subject of criticality safety and critical limits. Some of this literature is listed in Refs. 1–8. Reference 3 is an excellent and very readable report that covers the factors influencing critical parameters, critical limit data, computational techniques, and general criticality control practices. References 4 and 5 specify safety limits for a variety of conditions. References 6, 7, and 8 are three of the available compilations of experimental or calculated critical data.

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