

17 DATA ACQUISITION, REDUCTION, AND REPORTING

17.1 Introduction

This chapter provides information and guidance, primarily for laboratory personnel, on data acquisition, reduction, and reporting. Its intent is to provide an understanding of the many operational parameters which should be addressed in order that the data developed and reported are compliant with project planning documents (Chapter 4), considered valid (Chapter 8), and usable for their intended purposes (Chapter 9). These processes are all linked and each is dependent upon the results of its predecessor. The material presented is intended to provide an overview of the processes which are required in all radiochemistry laboratories, but are by no means performed in the same way in all laboratories.

In this chapter, data acquisition refers to the results produced by the radiation detection process, often referred to as counting. This chapter will provide guidance for laboratory personnel on selecting and applying the operational parameters related to instrumentation and the determination of the radioactivity contained in the test source.¹ Parameters that are applicable to counting for essentially all radiation detection instrumentation are discussed in Section 17.2 and those that are specific to a given type of instrumentation are covered in the appropriate section describing that instrument. A detailed description of the instrumentation discussed in this chapter was provided in Chapter 15.

Once test sources have been prepared (Chapter 16) and counted using laboratory measurement instruments (Chapter 15), the basic information generated by the instrument should be reduced (processed) to produce data which can be reviewed, verified, validated, and interpreted in light of and in accordance with project planning documents and analytical statements of work (SOWs) (Chapter 7). Data reduction is primarily mathematical in nature while data reporting involves the presentation of the results of the data acquisition and reduction processes and nonmathematical information necessary to interpret the data (e.g., sample identification and method of analysis).

Data reduction may be as simple as a division of the counts by the counting time, the sample aliquant weight or volume, and the counter efficiency, thereby producing the radionuclide concentration. On the other hand, it may also require more complicated processing such as the fitting of an analytical function, or the unfolding of a differential spectrum (Tsoulfanidis, 1983,

¹ The term “test source” will be used to describe the radioactive material prepared to be introduced into a measurement instrument and “laboratory sample” will be used to identify the material collected for analysis. Thus, a test source is prepared from laboratory sample material for the purpose of determining its radioactive constituents. “Calibration source” is used to indicate that the prepared source is for the purpose of calibrating instruments.

30 p. 327). In any case, the reduction process should continue by calculating the combined standard
31 uncertainty (Chapter 19).

32 The output of some laboratory instruments is highly simplistic and consists only of the number of
33 nuclear decay events recorded by the detector in the time interval allocated for the measurement.
34 An example of this might be a gas-proportional counter whose only output is an electronic scaler
35 and the available data consists of total counts or counts per minute. On the other extreme, some
36 laboratory counting instruments with computer components produce outputs consisting of
37 radionuclide concentration, uncertainty, and other information (see Chapter 19). Examples of
38 these types of data reducing instruments are alpha- and gamma-spectrometry and liquid-
39 scintillation systems.

40 ANSI N42.23 contains an outline of a minimal data report. Most project-specific planning
41 documents (Chapter 4) and/or analytical SOWs (Chapter 5) require that the radiochemical data
42 produced by laboratories be submitted in a specific format and form (i.e. electronic or hard copy,
43 or both). In some cases, the requirements are minimal and may consist of a data report which
44 gives only the sample identifier information, accompanied by the radionuclide concentration and
45 its associated uncertainty. Many projects require much more supporting information, primarily to
46 assist in the data validation (Chapter 8) process. Support material can include information on
47 calibration, background determination, sample processing, sample receipt, quality control sample
48 performance, raw-counting data, and chain-of-custody records.

49 This chapter gives an overview of data acquisition, reduction, and reporting in radiochemical
50 laboratories. The material presented is intended to be descriptive rather than prescriptive, since
51 these processes vary greatly between laboratories; depending upon the equipment, personnel,
52 project requirements, and the methods and analyses being performed.

53 **17.2 Data Acquisition**

54 Data acquisition refers to the process of collecting the basic information produced by nuclear
55 counting instruments. These data may be produced in hard copy or electronic format, or visually
56 displayed for the operator to record. As previously stated, this can be simply the number of
57 counts detected by the instrument within the allotted counting time or as conclusive as the
58 identification of the radionuclides contained in the sample along with their concentrations and
59 associated uncertainties.

60 Following generation, data requiring further processing may be electronically or manually
61 transferred to the next to the next data-reduction step. Electronic transfer should be employed as

62 often as possible to avoid the inherent errors associated with manual transfer. On the other hand,
63 the next step in the data reduction process may be performed manually, i.e., with a calculator.

64 The reliability of the data generated also depends upon the proper operation of the instrumenta-
65 tion and the associated data reduction programs. Data quality further depends upon the correct
66 input of associated information by laboratory personnel.

67 **17.2.1 Generic Counting Parameter Selection**

68 Instrument operators have choices, provided by instrument manufacturers, in the setup and
69 operation of nuclear counting instruments. These selections can affect the quality and
70 applicability of the data. Some selections can be made on a one-time basis and left unadjusted for
71 the processing of all samples and others require the operator to reevaluate the settings, possibly
72 for each test source counted. In some cases adjustments can be made following counting during
73 the processing of the derived information. Some adjustments can only be made before counting
74 or by extending the counting time. In making the proper selection, there are some overall
75 considerations relative to the project requirements, as specified in project planning documents
76 (Chapter 4) or in the analytical SOW (Chapter 5). Other operator decisions depend on the nature
77 of the test source itself. Caution should be exercised when changing operational parameters so
78 that the calibrations (counting efficiency, energy, self absorption, etc.) performed on the
79 instrument remain valid. For example, changing the source container or holder may affect the
80 counting efficiency and/or background. Determining the appropriate operating conditions
81 requires that the operator have a thorough understanding of the counting process and the
82 instruments and their operation for the production of valid and useable data. In addition, the
83 operator should be cognizant of the measurement quality objectives (MQOs) that have been
84 established.

85 Some of the factors that affect operational parameter selection are related to project requirements.
86 Planning documents and the analytical SOW may specify the limits on measurement uncertainty
87 and detection capability. In order to achieve compliance with the limits, instrument operating
88 parameter adjustment may be required for some or all the samples received. The number of
89 samples received during a time period may make it mandatory for adjustments to be made in
90 order to meet these requirements while complying with project defined turn-around-times.

91 Factors that may affect the selection of operational parameters include:

- 92 • Project and External
- 93 – project requirements for uncertainty, detection capability, and quantification capability

- 94 – laboratory backlog and contract turn-around times

- 95 • Sample Characteristics
- 96 – expected sample radionuclide concentration
- 97 – interfering radionuclides
- 98 – interfering stable constituents (e.g. liquid scintillation counting quenching)
- 99 – amount of sample available
- 100 – physical characteristics of the test source (e.g. density)
- 101 – half-life of the radionuclide of interest

- 102 • Analytical Process
- 103 – chemical separation process leading to counting source generation (Chapter 14)

- 104 • Instrumentation
- 105 – instrument adjustments available and their limits
- 106 – conditions and limits of an instrument’s calibration
- 107 – time availability of instruments
- 108 – counting efficiency
- 109 – calibration geometries available

110 Taking into consideration the above, the operator has control over and should select certain
111 parameters for all radiation measurements. The selection of the basic parameters should be
112 carefully planned in advance to assure that the project requirements are met. The laboratory’s
113 selection of parameters during the planning process may require alteration as the process of
114 sample analysis is actually taking place due to unavoidable changes in the samples and sample
115 characteristics throughout the duration of the study.

116 17.2.1.1 Counting Duration

117 For the Poisson counting model, the uncertainty associated with a given count determination is
118 proportional to the square root of the total number of counts accumulated (Chapter 19). The total
119 counts accumulated during counting are proportional to the activity of the source and the length
120 of the counting time. Counting duration is a controllable factor that allows one to achieve a given
121 level of counting uncertainty. The operator should then select a duration which is sufficient to
122 meet project objectives for detection capability and uncertainty. The length of time allotted for
123 determination of the instrument background will also affect the uncertainty associated with the
124 measurement (Chapter 19). Thus, when preparing an analytical protocol to meet the requirements
125 of a project, as expressed in the project planning documents, the laboratory will establish the

126 counting durations of both sample and background accordingly. An alternative to selecting a
127 counting duration, available on many instruments, is to count until a preset number of counts is
128 obtained.

129 17.2.1.2 Counting Geometry

130 The counting efficiency of a radiation detector depends on the geometry of the source and
131 detector arrangement, e.g., the solid angle subtended at the detector by the source. A given
132 radiation detector may have the counting efficiency established for several geometries. The
133 geometry selected among those available may depend upon the amount of sample available, the
134 detection capability required for the analysis, the radionuclide concentration in the sample, the
135 dictates of the radioanalytical method, the physical characteristics of the sample, the nature and
136 energy of the decay process, and the characteristics of the detector.

137
138 The choices to be made relative to geometry selection are usually the type of test source
139 container, the source mounting, and the detector to source distance. Choices are to be made
140 among those for which the detector has an established efficiency calibration.

141 17.2.1.3 Software

142 The use of properly developed and documented computer software programs for data acquisition
143 and reduction can lead to an enhancement in the quality of laboratory data. Guidance on software
144 documentation can be found in EPA (1995). Caution should be exercised in the selection and use
145 of undocumented programs and those which may not have been tested in laboratories performing
146 analyses similar to those for which MARLAP has been developed. For example, a spectral
147 analysis program may accurately identify and quantify the radionuclides in test sources
148 containing higher levels of radioactivity (which produce spectra with well defined peaks, easily
149 distinguishable from background) but may be inaccurate for samples with environmental levels.

150 When selecting software, a thorough review of the data reduction algorithms should be
151 performed. The user should not blindly accept the notion that all software performs the
152 calculations in an appropriate manner without this review. When evaluating software, it is often
153 helpful to review the software manual, particularly in regard to the algorithms used in the
154 calculations. While it may not be necessary that the user understand in detail all the calculations
155 performed by highly complex software programs, the user should understand the overall scheme
156 of analysis and reduction in order to assure data meet quality objectives and reporting
157 requirements. This understanding is also beneficial in assuring that user defined parameters are
158 properly selected.

159 The output of some instruments is very basic, primarily counting data, i.e., counts or counts per
160 second. These data should be manipulated by external systems to convert them to the form
161 required by planning documents. The external system which performs the calculations may be a
162 calculator or a computer with the appropriate software to reduce the data to usable terms. In
163 either case, additional information relative to the processing of the sample should be input along
164 with the counting data (counting time, total counts, and background counts). This information
165 may include laboratory sample number, collection date, sample mass or volume, instrument
166 counting efficiency, and chemical yield.

167 For computer (processor) based systems, some of this information is generated and processed
168 internally and the remainder is manually entered or electronically transferred from the Laboratory
169 Information Management System (LIMS) or some other adjunct system where it has previously
170 been stored. It is becoming increasingly common for much or all of this adjunct information to be
171 transferred to the counting instrument by reading a bar code affixed to the test source to be
172 counted. In this manner, the information which has previously been entered into a LIMS is
173 electronically transferred to the counting instrument. For hand calculations, these data are simply
174 entered into the calculations.

175 **17.2.2 Basic Data Reduction Calculations**

176 The equations used for data reduction depend on the analytical methods used. The following
177 equations are provided as examples to illustrate the basic principles involved in data reduction.

178 Following counting, the radionuclide concentration may be calculated:

$$R_C = \frac{C_{\text{Net}}}{\epsilon \cdot V \cdot Y \cdot K_C \cdot e^{-\lambda t_1}} \quad (17.1)$$

179 where:

- 180 R_C = radionuclide concentration at time of collection (Bq/L or Bq/g)
181 C_{net} = net count rate (cps)
182 ϵ = counter efficiency for the radionuclide (cps/dps)
183 V = volume or mass of sample analyzed (L or g)
184 Y = chemical yield (when appropriate)
185 e = base of natural logarithm
186 λ = the radioactive decay constant for the radionuclide (s^{-1} , min^{-1} , or d^{-1})

187 t_1 = time lapse from sample collections to beginning of source count (units consistent
188 with λ)
189 K_C = the correction for decay during counting and is:

$$K_C = \frac{1 - e^{-\lambda t_C}}{\lambda t_C} \quad (17.2)$$

190 where:

191 t_C = actual clock time (real time) of counting (units consistent with λ)

192 This calculates the radionuclide concentration at the time of sample collection². It compensates
193 for the fact that short-lived radionuclides may experience significant reduction in activity during
194 counting, when the counting duration is a significant fraction of the half-life. For long-lived
195 radionuclides, the term K_C approaches unity and can be ignored. The efficiency used in this
196 equation may be obtained from the specific radionuclide whose concentration, R_C , is to be
197 determined or it may be obtained from an efficiency curve which plots counter efficiency vs.
198 energy. In the latter case, the abundance, E_e , of the particle or photon being counted should be
199 considered. This is required because the energy dependent efficiency, ϵ_e , is developed in terms of
200 the fraction of particles or photons detected divided by the number emitted at that energy. Thus,
201 if the radionuclide emission being determined during the counting of a test source has an
202 abundance less than 100 percent, an adjustment should be made to Equation 17.1, as shown in
203 Equation 17.3:

$$R_C = \frac{C_{\text{Net}}}{E_e \cdot \epsilon_e \cdot V \cdot Y \cdot K_C \cdot e^{-\lambda t_1}} \quad (17.3)$$

204 Most modern instrument systems contain preprogrammed software to perform data manipula-
205 tions that convert basic counting information to a form which can be compared to the project data
206 quality objectives, or at least to begin or promote this process. Certain sample-specific
207 information should be manually entered or transferred to the system electronically in order to
208 perform the necessary calculations.

² For radionuclides with short half-lives detected at or near detection limits, it may be more appropriate to calculate the concentration at the time of counting.

209 **17.3 Data Reduction on Spectrometry Systems**

210 Software is available for resolving alpha, gamma, and liquid scintillation spectra and for
211 performing the attendant functions such as calibration, energy alignment, background acquisition
212 and subtraction, and quality control functions.

213 Spectroscopic analysis for alpha particles and gamma-rays is performed to identify and quantify
214 radionuclides in samples. Since these emissions occur at discrete energies, spectrometry is useful
215 for these purposes and can be applied to the analysis of a wide range of radionuclides. Energy
216 spectra are produced when a detector absorbs a particle or photon and produces a signal that is
217 proportional to the energy absorbed. The resulting signal is digitized by an analog-to-digital
218 converter and processed by a multichannel analyzer. A differential spectrum is produced, where
219 the number of events within an incremental energy, ΔE , is recorded on the y axis and the energy
220 is represented on the x axis (Tsoulfanidis, 1983, p. 327). In this way, radionuclides can be
221 identified by the characteristic energies of their emissions and quantified because the area under
222 the full energy peak is proportional to the emission rate (activity) of the source being analyzed.

223 The spectra for alpha and gamma emitters are quite different, due to the differences in the way
224 these two types of radiation interact with matter in transferring their energy to the detector
225 material. The process of resolving the spectra into its contributing components is referred to as
226 spectral analysis (NCRP 1978, p. 159) and unfolding (Tsoulfanidis, 1983, p. 342). Computer
227 programs for analyzing alpha and gamma spectra are available from several sources (Decker and
228 Sandderson, 1992). A method of performance testing of gamma analysis software is given in
229 ANSI N42.14.

230 **17.3.1 Gamma Spectrometry**

231 Gamma spectrometry on environmental samples requires the use of gamma spectral analysis
232 software for any reasonable degree of accuracy and detection capability. This is due to the
233 potentially large number of photopeaks to resolve, the low level of radioactivity in most
234 environmental samples, and the relatively low detection limits and stringent quality control
235 requirements of most project-specific planning documents. Spectral analysis by manual
236 techniques is only practical when the number of radionuclides is limited and the contributing
237 isotopes are predictable. An example is the analysis of milk samples for gamma-emitting
238 radionuclides, where the milk production process in the cow restricts the number of radionuclides
239 in the milk product (Hagee et al., 1960, p. 36; USPHS, 1967, pp. 1–51).

240 Gamma-rays interact with matter in
 241 three ways, namely, by photoelectric
 242 effect, Compton scattering, and pair
 243 production (Tsoulfanidis, 1983, pp.
 244 141–148). These interactions within a
 245 gamma detector, usually a high-purity
 246 germanium or sodium iodide (see
 247 Chapter 16), result in varying amounts
 248 of the gamma-ray energy being
 249 absorbed. Only one of these inter-
 250 actions, the photoelectric effect, results
 251 in the total energy being absorbed in a
 252 single interaction. The photopeak,
 253 shown in Figure 17.1, due to a
 254 photoelectric interaction in the detector,
 255 results from the processing of the
 256 detector signal through the linear
 257 circuitry and the multichannel analyzer.

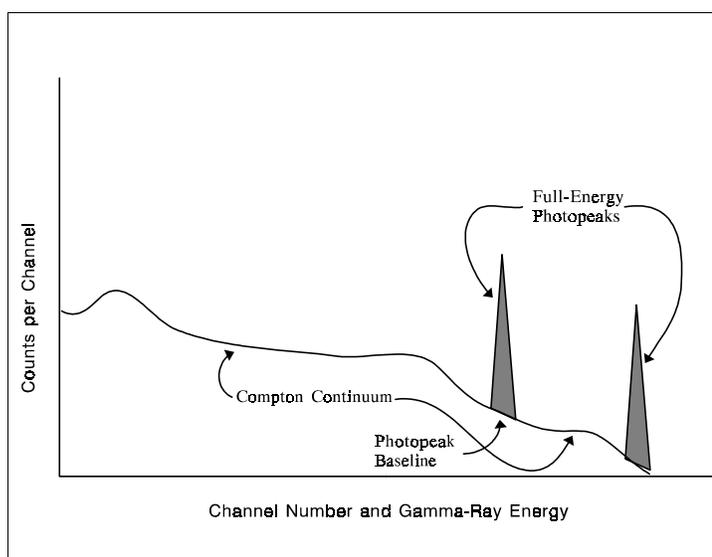


FIGURE 17.1 — Gamma-ray spectrum

258 This photopeak has a basic Gaussian shape (Gilmore and Hemmingway, 1995, p.163) and may
 259 be described by (Quittner, 1972, p.20):

$$y(x) = A e^{-(x-p)^2 / 2\sigma^2} \quad (17.4)$$

260 where:

261 A = the peak amplitude
 262 x = the channel number
 263 p = the peak centroid

264 (The width of the peak is related to the full-width at half-maximum (FWHM) of the detector, Γ ,
 265 where $\Gamma = 2.355 \sigma$. The area under the peak is $N = 1.064 A \Gamma$.)

266 As can be seen in Figure 17.1, the photopeak (P1) may be displaced upward by its position on the
 267 Compton continuum from other, higher-energy gamma-rays (P2) and background radiation.

268 The photopeak is the key element in gamma-ray spectrometry in that its location on the energy
 269 axis provides a means for radionuclide identification, and the area under the peak is proportional

270 to the number of gamma-ray events comprising the photopeak. This becomes the basis for
271 radionuclide identification and quantification.

272 The fundamental purposes of gamma-ray computer-based spectral analysis programs are to
273 identify the photopeaks in a spectrum and to measure the true area under the photopeaks. It
274 should do this in the presence of natural background, a potentially large number of sometimes
275 overlapping photopeaks, and a great number of Compton-scattering events. Once these initial
276 tasks have been performed, the computer program uses this information to determine the
277 radionuclide mix that contributed the complex spectrum and the individual concentrations in the
278 sample being analyzed.

279 Most computer programs for gamma-spectral analysis are provided by equipment manufacturers,
280 although some are supplied by independent providers. There are significant differences in the
281 structure of the programs. However, they all perform similar functions which are given below
282 and illustrated in Figure 17.2.

283 17.3.1.1 Peak Search or Identification

284 There are two basic methods of gamma spectral analysis. The first method is to allow the
285 analysis software to determine the existence of the peaks and their energy. The second method is
286 often referred to as a “library directed” search, where the operator identifies the peak energy
287 locations, e.g., regions of interest, to be searched for discernable peaks. The latter method may be
288 more sensitive (Gilmore and Hemmingway, 1995, p.165) but, taken alone, will fail to identify
289 and report unspecified radionuclides. If the confirmation of the existence of a particular
290 radionuclide is required, the second method should be employed. Most software programs allow
291 either approach to be activated and used for each analysis.

292 A most important function performed by an analysis program is the identification of true
293 photopeaks. In the programs available, this is achieved in one of the four ways discussed below.

294 Many spectral analysis programs allow the operator to select among two or more of the four
295 methods for peak identification. Selection of the most accurate and sensitive method depends on
296 the radionuclides present in the source, detection capability requirements for individual
297 radionuclides, the number of radionuclides present, the nature of the background spectrum, the
298 degree to which the radionuclide mix can be predicted, and the activities of the isotopes. The
299 selection of a particular peak search method can be determined by experience with similar
300 sample types and past performance, particularly on performance evaluation (known) samples.

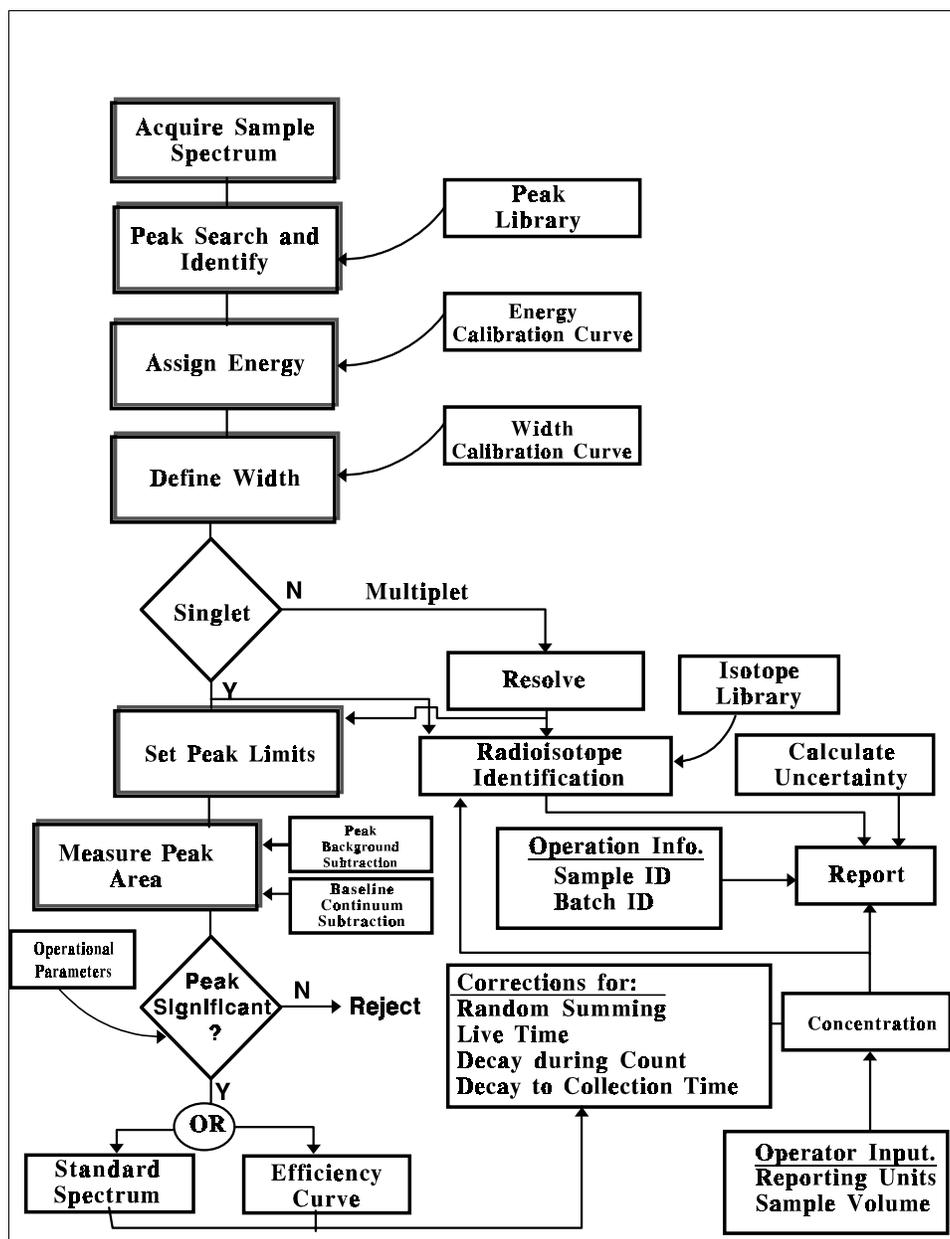


FIGURE 17.2 — Gamma-ray analysis sequence

301 REGIONS OF INTEREST (ROI) METHOD

302 This is the simplest form of peak identification, but can only be used when the radionuclides

303 present in the sample are known and when the analysis system has been compensated for gain
304 drift. ROI analysis involves the establishment of predetermined energy regions, at least one for
305 each radionuclide present. Once the spectrum has been acquired, the number of counts in each
306 region is summed after subtracting the photopeak baseline (Figure 17.1). This method of spectral
307 analysis is more applicable to alpha rather than gamma spectrometry.

308 GAUSSIAN FUNCTION DERIVATIVE METHOD

309 As previously stated, the photopeak has a basic Gaussian shape; in reality it is a histogram with a
310 Gaussian-like shape. The most widely used peak identification technique was proposed by
311 Mariscotti (Mariscotti 1967, p. 309) and uses the Gaussian function derivative to assess the
312 presence of a photopeak. For most low-level radioactivity, this peak search method may provide
313 the best peak detection capability with the fewest false peak identifications or omissions of true
314 peaks (Gilmore and Hemmingway, 1995, p. 20).

315 CHANNEL DIFFERENTIAL METHOD

316 This method searches for a number of channels where the counts are significantly greater than the
317 preceding channels, and then looks for the expected decrease in counts corresponding to the
318 backside of the prospective photopeak. This method works relatively well for large, well-defined
319 peaks, but is limited for poorly defined peaks with counts barely above the background baseline
320 of the peak (Gilmore and Hemmingway, 1995, p. 163).

321 CORRELATION METHOD

322 In this method, a search function is scanned across the spectrum. Each channel count, over the
323 width of the search function, is multiplied by the corresponding value of the search function. The
324 sum of these products is then made a point on a correlation spectrum. A correction for the
325 baseline contribution leaves only positive counts within a photopeak. Although the scan function
326 is normally Gaussian in form, other forms may be applied (Gilmore and Hemmingway, 1995,
327 p. 164).

328 Spectral analysis programs usually have some user selected peak acceptance criteria. The
329 acceptance criteria may be based on peak shape, width uncertainty, or the number of standard
330 deviations above the background to be subtracted. Care is required in selection of the values for
331 these acceptance criteria. If the values are too high, valid photopeaks remain undetected. If the
332 values selected are too low, radionuclides may be reported which are not present in the samples.
333 Knowledge of the sample origin and experience with using the analysis program on similar

334 samples to those being processed is useful in establishing values for these user-selected
335 parameters. Peak searches may be standard or directed (Canberra, 1994). In a standard search, all
336 peaks identified are assigned to a library contained radionuclide. In a directed search, the user
337 specifies the energies and radionuclides over which the search is performed. If reporting of a
338 specific radionuclide is required, the directed search is appropriate; however, some radionuclides
339 could go unreported if only a directed search is performed.

340 17.3.1.2 Singlet/Multiplet Peaks

341 A peak is referred to as a singlet or multiplet according to whether it is composed of a single
342 photopeak or multiple photopeaks, respectively. Deconvolution is the term given to the process
343 of resolving a multiplet into its components (Gilmore and Hemmingway, 1995, p. 172). The
344 ability of a spectral analysis program to perform this function may well be the deciding point for
345 its selection. It is particularly important if the laboratory has analyses in which one of the critical
346 radionuclides has only one gamma-ray whose energy is very near to that of another radionuclide
347 expected to be present in all or most samples.

348 There are three primary ways that programs deal with the problem of resolving multiplets. The
349 first method is a deconvolution algorithm which is based on the peak-shape being the composite
350 of multiplet Gaussian distributions. The second method uses the gamma-ray library to anticipate
351 where peaks occur within a multiplet. The disadvantage of the first is in dealing with small ill-
352 defined peaks and the second cannot, of course, resolve peaks not included in the library. The
353 third method, peak stripping, again depends on defining all radionuclides whose gamma-rays
354 contribute to the multiplet. In peak stripping, one of the interfering gamma-ray's contribution is
355 subtracted from the multiplet area by using another of its gamma-rays to estimate the peak shape
356 and size in the multiplet area. The remaining peak is, presumably, that of the interfered
357 radionuclide which can then be identified and quantified. This method requires that one of the
358 interfering radionuclides have a second gamma emission which identifies and tentatively, for the
359 purpose of removing its contribution, quantifies it.

360 In some cases, the uncertainty of multiplet deconvolution can be avoided by selecting photopeaks
361 from gamma-rays which are not interfered with, even though they may have lower abundances.
362 The increase in uncertainty due to the lower number of accumulated counts may well overcome
363 the uncertainty of deconvolution (Gilmore and Hemmingway, 1995, p. 174).

364 17.3.1.3 Definition of Peak Centroid and Energy

365 Once a peak has been detected, the centroid of the peak will be defined, since it will rarely be
366 located at exactly a whole channel number. The centroid will be used to represent the gamma-ray
367 energy and should be calculated to the fraction of a channel. An algorithm is used to calculate the
368 centroid value may be expressed as (Gilmore and Hemmingway, 1995, p. 167):

$$\text{Centroid} = \frac{\sum C_i i}{\sum C_i} \quad (17.5)$$

369 where:

370 C_i is the count in the i^{th} channel.

371 In order to assign a gamma-ray energy value to the peak centroid channel position, the analysis
372 program refers to a previously established energy calibration file. The detector's response to the
373 full range of gamma energies should be established by counting a source(s) having a number of
374 well-defined gamma-rays over the range of energies emitted by the radionuclides in the
375 calibration source. This calibration source is most often a "mixed-nuclide source," which also
376 has certified emission rates so that it may also be used for an efficiency calibration. The mixed-
377 nuclide source is counted on the detector, being sure to accumulate sufficient counts in the peaks
378 to obtain good statistical precision, and an energy-versus-channel relationship is established. The
379 operator will be required to provide information on the peaks to be used and their exact energies.

380 With modern spectrometry systems, the relationship between energy and channel number is
381 nearly linear. Both linear and quadratic fits have been included in available spectral analysis
382 programs.

383 17.3.1.4 Peak Width Determination

384 In order to calculate the area under the peak, an estimate of the peak width is required, unless the
385 analysis program is operating in the region-of-interest mode. The width of a photopeak is
386 normally quoted in terms of its FWHM. For a discussion of peak width (resolution) and the
387 factors affecting it, see Chapter 15.

388 There are several ways to determine the peak boundary. These are:

- 389 (1) A Gaussian shape is assumed and some number of standard deviations (2 or 3) are
390 allowed on each side of the peak centroid.

391 (2) A standard width for each peak, based on its energy, is used.

392 (3) A five-point moving average is used to determine a minimum on each side of the peak,
393 which is set as the peak limits.

394 Each method has strengths and weaknesses, but all struggle with ill-defined (small number of
395 counts) peaks. Once the peak limits are defined, determining the area under the peak is
396 accomplished by summing the counts per channel for the channels contained in the peak and
397 subtracting the baseline (see Figure 17.1).

398 The determination of FWHM requires an assumption of peak shape and, as has previously been
399 stated, the acceptance of a Gaussian function is the norm for gamma spectrometry. In addition,
400 the peak width increases with the energy of the gamma-ray, so some function should be defined
401 for the analysis program to determine the width based on the energy of the peak. This
402 relationship, in practice, is found to be nearly linear (Gilmore and Hemmingway, 1995, p. 133)
403 and described by:

$$w = a + bE \tag{17.6}$$

404 where:

- 405 w = width of the peak
- 406 E = the energy
- 407 a, b = empirical constants

408 For spectra developed by high-purity germanium semiconductors (HPGe) and alpha solid state
409 detectors, it is more appropriate to assume a peak shape which is a modification of the Gaussian
410 function to allow for the low energy tailing observed in these spectra. This type of tailing is
411 illustrated in Figure 17.3. Some spectroscopy programs have algorithms to fit peaks with lower
412 energy tailing.

413 When the “tailing” peak fit option is selected, the software algorithm for peak fitting changes
414 from the pure Gaussian form to a dual fit. The channels in the peak not affected by the tailing are
415 included in the Gaussian fit (Equation 17.7), and those that are affected by tailing are modified
416 according to Equation 17.8, below:

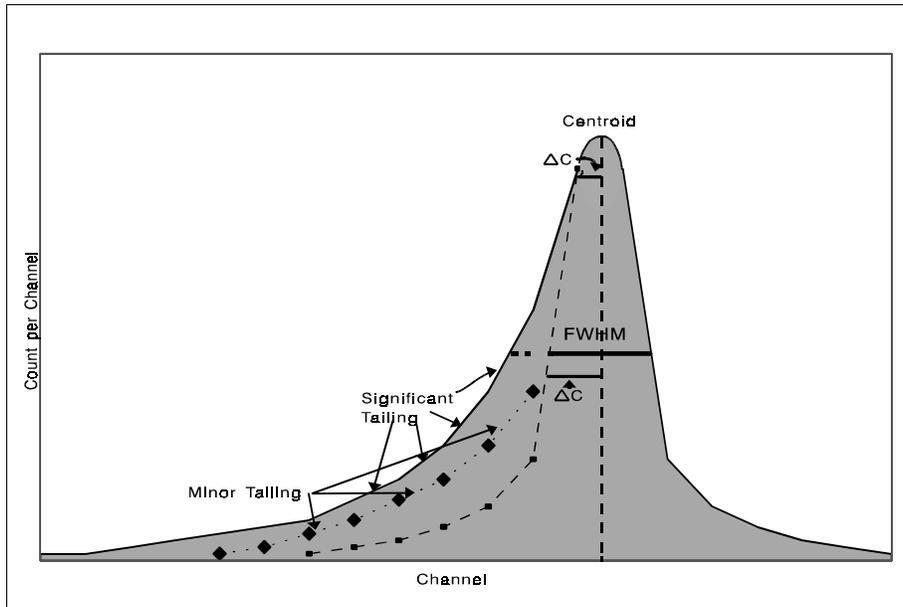


FIGURE 17.3 — Low-energy tailing

$$y(x) = \begin{cases} A e^{-\frac{(x - P_C)^2}{2\sigma^2}}, & x \geq P_C - \Delta C \\ A e^{\frac{\Delta C(2x - 2P_C + \Delta C)}{2\sigma^2}}, & x < P_C - \Delta C \end{cases} \quad (17.7)$$

$$A e^{\frac{\Delta C(2x - 2P_C + \Delta C)}{2\sigma^2}}, \quad x < P_C - \Delta C \quad (17.8)$$

417 where:

418 x = the channel number

419 A = the peak amplitude

420 P_C = the peak centroid

421 ΔC = the tailing factor (the distance from the centroid to the point where the tailing
422 joins the Gaussian peak)

423 σ = the width of the Gaussian peak ($\approx 2.355 \times \text{FWHM}$)

424 17.3.1.5 Peak Area Determination

425 For single peaks sitting on a Compton continuum, two methods of peak area determination are
 426 available. The simpler method is the addition (integration) of the number of counts per channel in
 427 each of the channels considered to be within the peak limits, and subtracting the natural
 428 background and Compton contribution to those same channels (Baedeker, 1971; Loska, 1988).
 429 However, this is rarely simple since the photopeak is usually offset by a baseline continuum
 430 whose contribution is not easily determined. While the background may be subtracted by the
 431 spectrometry program, the Compton continuum will be estimated by the software and then
 432 subtracted. This estimation is often based on the number of counts per channel in those channels
 433 immediately above and below the photopeak region as shown in Figure 17.4.

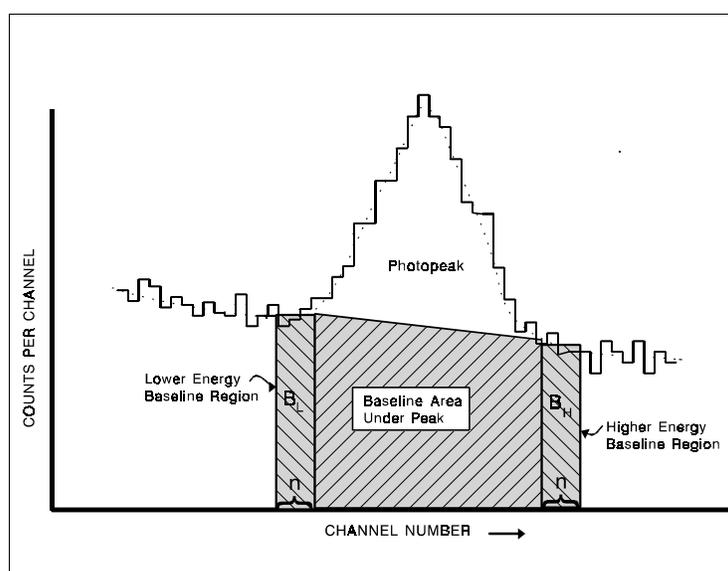


FIGURE 17.4 — Photopeak baseline continuum

434 The baseline contribution is then estimated as:

$$B = \frac{N}{2n} (B_L + B_H) \quad (17.10)$$

435 where:

436 B = the number of counts attributed to the baseline

437 N = number of channels in the peak

- 438 n = the number of baseline channels considered on each side of the peak for calculating
 439 B_L and B_H
 440 B_L = the sum of the number of counts in the baseline region on the low-energy side
 441 B_H = the sum of the number of counts in the baseline region on the high-energy side

442 In practice, the baseline continuum appears to have a step beneath the peak (Gilmore and
 443 Hemmingway, 1995, p.114), as illustrated in Figure 17.5. This type of function is estimated by:

$$B = \sum_{i=1}^N \left[\frac{B_L}{n} + \frac{B_H - B_L}{nG} \sum_{j=1}^i y_j \right] \quad (17.11)$$

444 where:

- 445 B_L = sum of counts in the baseline region on the low-energy side
 446 B_H = sum of counts in the baseline region on the high-energy side
 447 y_j = counts per channel in channel j
 448 G = gross counts in the peak
 449 N = number of channels in the peak
 450 n = number of channels in each of the two baseline regions

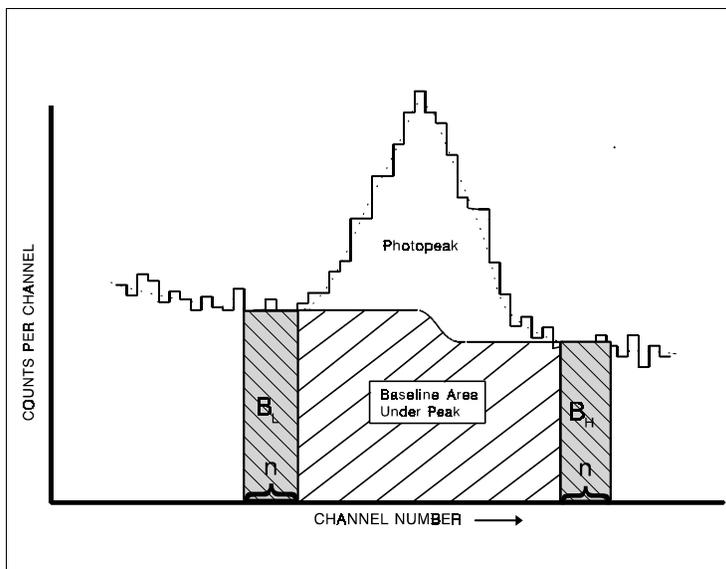


FIGURE 17.5 — Photopeak baseline continuum-step function

451 The second peak area determination method is the least-squares method, which fits a theoretical
452 peak shape plus background shape to the channels surrounding the peak (Kruse and Spettel,
453 1982; Helmer et al., 1983). Background is often subtracted prior to the fitting process (Loska and
454 Ptasiński, 1994).

455 17.3.1.6 Calibration Reference File

456 Three types of calibrations are required for gamma spectral analysis, namely those for efficiency,
457 energy, and FWHM. Efficiency and energy calibrations require a source whose gamma-ray
458 emission rate is known and referenced to a national standard, and whose gamma-ray energy lines
459 are well known. “Mixed radionuclide” reference material, containing eight or more gamma lines,
460 is available for performing these spectral calibrations. The operator is required to enter the
461 pertinent information, usually listed in the calibration source certificate, into the file prior to
462 performing the calibrations. The information generally consists of:

- 463 • Radionuclide name;
- 464 • Certified activity and units;
- 465 • Uncertainty in activity;
- 466 • Reference date and time;
- 467 • Gamma energies and branching ratios; and
- 468 • Half-life.

469 Once calibration files are established, the calibrations are performed according to methods
470 specific to individual software and as described in manufacturers manuals (also see Chapter 16).

471 17.3.1.7 Activity and Concentration

472 In order to convert the counts under a photopeak to activity, an efficiency calibration should be
473 performed on the detector. Since the efficiency varies with energy, the detector should be
474 calibrated over the range of energies to be used and a calibration curve developed for the
475 detector. In constructing an efficiency calibration curve, only calibration sources with singlet
476 peaks and well-known abundances should be selected. The efficiency, at a specific energy, is
477 simply the number of counts determined in a photopeak of known energy divided by the number
478 of gamma-rays emitted by the source in the same time period, or:

$$\epsilon = \frac{C_r}{D} \quad (17.12)$$

479 where:

480 ϵ = efficiency in cps/ γ ps

481 C_r = cps in the photopeak

482 D = gamma emission rate of source in dps

483 The efficiency versus energy curve developed in most gamma software packages is in the form of
484 a polynomial. One such form is:

$$\ln \epsilon = \sum_{i=0}^n b_i \cdot [\ln E]^i \quad (17.13)$$

485 where:

486 ϵ = full peak efficiency

487 b_i = coefficient as determined by calculation

488 E = the energy of the photopeak

489 The efficiency curve for high-purity germanium detectors shows two distinctive slopes. The
490 polynomial fit in some analysis programs allows for a dual fit, i.e., a separate fit is made to the
491 two portions of the curve.

492 This efficiency curve is maintained in the calibration file of the spectral analysis program to be
493 applied to each analysis. An efficiency curve should be maintained for each test-source geometry
494 to be used for the calibrated detector.

495 To obtain the activity in the test source, the net counts (background subtracted) in the photopeak,
496 as determined by the software through the process described above, is divided by the geometry-
497 specific efficiency. The activity units are converted to those selected by the operator and
498 corrected for decay to the time of collection. Based on sample-aliquant size/volume information
499 supplied by the operator, sample concentration is calculated and reported.

500 17.3.1.8 Summing Considerations

501 Summing refers to the summing of the energy of two or more gamma-rays when they interact
502 with the detector within the resolving time of the spectrometer's electronics. There are two types
503 of summing: (1) *random summing*, where two unrelated gamma-rays are detected at the same
504 time, and (2) *true coincidence summing*, is due to the simultaneous emission of gamma-rays by a
505 radionuclide and their subsequent detection by the gamma detector.

506 Random summing, sometimes referred to as pile-up, is due to gamma-rays, from different
507 radionuclides, being detected almost simultaneously. If two gamma-rays arrive at the detector
508 within the resolving time of the amplifier and both have a photoelectric interaction, instead of
509 having a count in both full-energy peaks a count will occur somewhere else in the spectrum equal
510 to the sum of the two energies. Random summing can also occur with other than photoelectric
511 interactions, e.g., photoelectric with Compton and Compton with Compton. Since this occurs
512 randomly in nature, the probability of random summing increases with the square of the total
513 count rate. Random summing can be reduced by the use of pile-up rejection circuitry which
514 examines the pulse shape of detector signals and rejects those which are distorted by summing
515 (Gilmore and Hemmingway, 1995). However, even with pile-up rejection random summing will
516 still be present. A mathematical correction for random summing is given by:

$$A_T = A e^{2R\tau} \quad (17.14)$$

517 where:

518 A_T = the true peak area (counts)
519 A = the observed peak area (counts)
520 R = the mean (total) count rate (cps)
521 τ = the resolving time of the electronics (μ s)

522 If unknown, the resolving time can be estimated by a method similar to that described in Gilmore
523 (1995).

524 True coincidence summing is a source of error when a source contains nuclides which emit
525 gamma-rays nearly simultaneously. Coincidence summing is geometry dependent and increases
526 as the source is positioned closer to the detector. Thus, the use of multi-gamma-ray calibration
527 sources for close geometry efficiency calibrations must be done with caution. True coincidence
528 summing also increases with detector volume and is very prevalent in a well detector. The use of
529 a detector with a thin entry window opens the possibility of coincidence summing with X-rays.
530 Since coincidence summing is independent of count rate, it is a mistake to assume that the
531 measurement of environmental media is immune from errors caused by this phenomena.

532 As is the case with random summing, true coincidence summing results in the loss of counts
533 from photopeaks and a corresponding loss in efficiency. The use of single gamma-ray emitting
534 radionuclides is recommended, to the extent possible, for developing calibration curves for
535 detectors at close geometries. In practice, even when the efficiencies are determined in this
536 manner, errors in analyzing for nuclides emitting more than one gamma-ray still exist. When a
537 multi-emitting gamma-ray source is to be measured with minimum bias, it may be necessary to

538 perform an efficiency calibration with the specific radionuclide to be measured in the specific
539 geometry desired.

540 In theory it is possible to mathematically correct for true coincidence summing; however, for
541 complicated decay schemes, the task is daunting (Gilmore and Hemmingway, 1995). Some data
542 have been published which give correction factors for coincidence summing for a number of
543 radionuclides (Debertin and Helmer, 1988). Unfortunately they only apply to the particular
544 detector and geometries for which they were developed.

545 17.3.1.9 Uncertainty Calculation

546 The various components of uncertainty in the determination of the source activity should be
547 propagated to obtain the combined standard uncertainty. The sources of uncertainty in the gamma
548 spectral analysis include those associated with the determination of the net peak area, which
549 includes the standard uncertainties of the gross counts, the background counts, and any
550 interference from other gamma radionuclides present; the uncertainty associated with the
551 unfolding of multiplets; the detector efficiency, which includes uncertainties of the net peak area,
552 the calibration source emission rate, and decay correction factor; and uncertainty in the
553 determination of the sample volume or mass.

$$u_c = \sqrt{u_p^2 + u_v^2 + u_e^2 + u_U^2} \quad (17.15)$$

554 where:

555 u_c = the combined standard uncertainty

556 u_p = the component of combined standard uncertainty due to the net peak area
557 determination

558 u_v = the uncertainty component for the volume or mass determination

559 u_e = the uncertainty component for the efficiency determination

560 u_U = the uncertainty component for the unfolding routine for multiplets

561 Each of these factors may have a number of components of uncertainties included, for example,
562 the net peak uncertainty:

$$u_p = \sqrt{u_G^2 + u_B^2 + u_E^2 + u_I^2} \quad (17.16)$$

563 where:

564 u_G = the uncertainty component for the gross counts in the peak

565 u_B = the uncertainty component for the baseline subtraction
566 u_E = the uncertainty component for the background peak subtraction
567 u_I = the uncertainty component for the coincidence summing correction

568 The calculations of combined standard uncertainty typically are performed by the spectrometry
569 software for an alpha-spectrometry analysis. It should be noted that not every available software
570 package will incorporate all the listed uncertainty contributions listed.

571 **17.3.2 Alpha Spectrometry**

572 This section deals with alpha spectrum reduction as applied to semiconductor detectors, since it
573 is likely that this is the type of detector that will be employed for environmental analyses.

574 Since the range of alpha particles is a few centimeters in air and their energy is significantly
575 degraded in passing through a few millimeters of air, alpha spectrometry is conducted in a partial
576 vacuum and on extremely thin sources prepared by electrodeposition or coprecipitation (see
577 Chapter 16).

578 The number of full energy peaks is usually not large, three to four, in an alpha spectra and they
579 are normally well separated in energy. This, coupled with the fact that the test source subjected to
580 counting has gone through a chemical separation (Chapter 14), makes the radionuclide identifica-
581 tion relatively simple when compared to gamma spectrometry. However, it is still of great benefit
582 to have alpha spectrometry software to identify s radionuclides, subtract background, perform
583 calibrations and energy alignments, determine radiochemical yields, and perform and track
584 quality control functions. In production laboratories where hundreds of alpha spectra may be
585 generated each week, it is almost imperative that alpha spectra are resolved by properly designed
586 computer software. An alpha spectrum produced by a semiconductor detector by the counting of
587 a thin source containing ^{234}U , ^{238}U , ^{239}Pu , and ^{241}Am is shown in Figure 17.6.

588 The spectrum demonstrated contains
589 four peaks which are distorted from
590 their basic Gaussian shape because
591 each of the isotopes emits more than
592 one alpha particle whose energies
593 are within the resolving power of the
594 detector and electronics. The
595 FWHM of the peaks shown is
596 approximately 30 keV. Of particular
597 note is the fact that the peaks are
598 essentially sitting on the baseline.

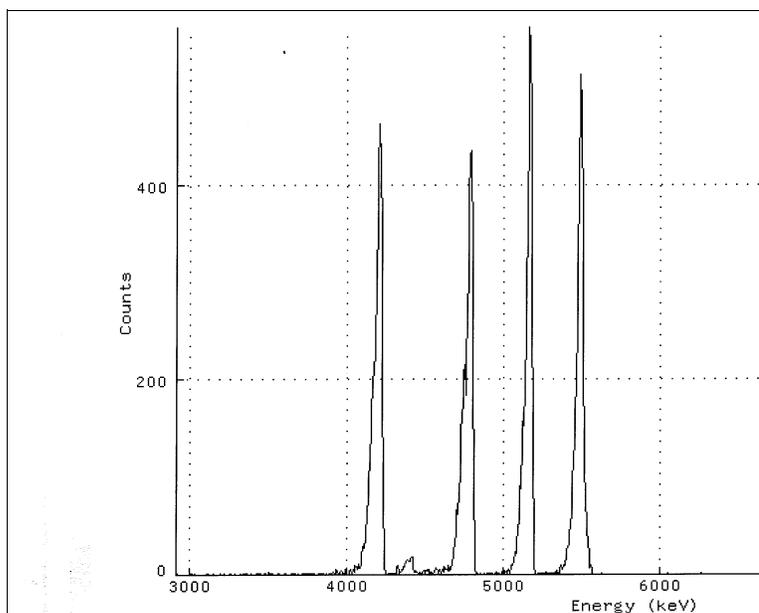


FIGURE 17.6 — Alpha spectrum

599 Spectral analysis programs usually
600 have routines for the identification
601 of full-energy peaks. However, in
602 the case of alpha spectrometry,
603 because the locations of peaks in the
604 spectrum are known and the peaks
605 may contain a small number of

606 counts, an ROI-type of analysis is usually performed. However, peak fitting programs are
607 available and may be beneficial when overlapping of peaks is possible. The algorithms used for
608 peak fitting of alpha spectra should take into account the low energy tailing present in most alpha
609 sources (Equation 17.8). The algorithms which account for tailing are modified Gaussian
610 functions and require a peak shape calibration where a number of well-defined singlet peaks
611 covering the full energy range are acquired. The calibration program then calculates the tail
612 parameter values (see discussion on tailing in Section 17.3.1.4, “Gamma Spectrometry”).

613 Alpha peaks are normally sitting on the baseline (no background continuum) and display
614 minimal overlapping for well-prepared sources. For a given analysis (Pu, U, Am, Th, and etc.),
615 ROIs are established for all energies of the alpha emissions in the source being counted and the
616 count rate in a given ROI represents the emission rate of the alpha whose energy falls within that
617 ROI.

618 Given these qualifications, the spectral analysis software performs essentially the same functions
619 as that for gamma analysis, described above. The programs may also perform system control
620 function, e.g., maintaining vacuum in the chambers. Databases related to procedures, chemical
621 tracers, and efficiency and energy calibration standards are normally maintained for calculational,

622 documentation, and quality control purposes. The general analysis sequence for alpha
623 spectrometry will be briefly discussed below.

624 An efficiency calibration is not an absolute necessity if a standard/reference material is used for a
625 tracer in each sample and an accurate determination of the yield is not required. In some cases,
626 the laboratory may perform an energy and efficiency calibration for an alpha spectrometry
627 analysis. This requires the operator to establish a calibration certificate file for the program to
628 reference. It should refer to this file for both energy and efficiency calibrations. Calibration
629 sources are necessary for performing the required calibrations, and the appropriate certificate
630 information should be entered into the certificate files in order to perform the calibrations and to
631 analyze test sources. This information should be supplied with calibration sources. Calibration
632 sources, consisting of three to four radionuclides, are available in the form of plated discs from
633 several commercial suppliers.

634 Information typically required by the analysis program consists of the following:

- 635 • Radionuclide
- 636 • Activity
- 637 • Assay date
- 638 • Half-life
- 639 • Energy
- 640 • Energy uncertainty
- 641 • Emission probability per event
- 642 • Emission rate uncertainty
- 643 • Activity units

644 This information should be entered for each of the radionuclides included in the calibration
645 source. Once the library file has been established, an energy calibration can be performed as
646 directed by the software program.

647 The efficiency for alpha particles varies only slightly with energy, within the range of alpha
648 energies usually encountered. While the calibration source may contain several certified
649 radionuclides, during an efficiency calibration, the mean efficiency for the full-energy peaks may
650 be calculated and used as the alpha efficiency for a given detector (Chapter 16).

651 Once the alpha spectrometry system has been calibrated and a spectrum of a test source acquired,
652 either a peak search is performed to identify alpha peaks or, if operating in a ROI mode, the
653 counts in the ROI are determined. ROIs to be used for a given analysis are established prior to the

654 spectrum acquisition by selecting an analysis protocol where the radionuclides and their alpha
655 energies are preestablished.

656 In the ROI mode, the counts accumulated during the preset counting duration in each of the
657 designated regions are corrected for background contribution and, in some cases, for reagent
658 blank activity. If a tracer has been added to the test source, the counts in the tracer ROI are
659 summed, background corrected, and the effective efficiency (yield times counting efficiency)
660 determined using certificate information previously entered by the operator and/or from a
661 protocol file. The yield, if required, is then computed by the use of an efficiency which has been
662 previously determined during an efficiency calibration process. The radionuclide concentration is
663 then calculated by³:

$$R_{C_i} = \frac{C_{R_i}}{\epsilon_e \cdot V \cdot e^{-\lambda_i t_1}} \quad (17.17)$$

664 where:

665 R_{C_i} = radionuclide concentration of the radionuclide at time of collection (Bq/L or Bq/g)
666 C_{R_i} = net count rate in the designated ROI for the radionuclide (cps)
667 ϵ_e = effective efficiency ($\epsilon \cdot Y$) for the tracer (cps/dps)
668 V = volume or mass of sample analyzed (L or g)
669 e = base of natural logarithm
670 λ_i = the radioactive decay constant for the radionuclide (s^{-1} , min^{-1} , or d^{-1})
671 t_1 = time lapse from sample collection to beginning of source count (units consistent
672 with λ_i)

673 Following the spectrum acquisition process, spectral analysis programs may either automatically
674 process the data and present the results, or they may store the spectral data and await interaction
675 from the operator for processing. In either case, post-acquisition review of the analysis results is
676 recommended. This review may include the following items:

- 677
- 678 • Assure that the alpha peaks fall within the ROIs;
 - 679 • Confirm the absence of unexpected peaks (contamination);
 - 680 • Verify that there are no interfering peaks;
 - 681 • Confirm that peak centroids are within requirements (energy alignment);
 - Verify that all requirements are met with regard to FWHM and chemical yield; and

³ For certain alpha-emitting radionuclides, ²²⁴Ra for example, a decay-correcting term is needed.

- 682 • Check units and sample aliquant information.

683 The FWHM of a given peak may depend greatly on the source preparation. However, since an
684 ROI-type of peak search is normally used, and the limits of the peak determined by the setting of
685 the ROI rather than some algorithm, the peak width definition is not significantly affected by
686 reasonable peak broadening. As a precautionary measure, the above review of each test-source
687 spectrum assures that the peaks appear within the ROIs. Alpha spectrometry analysis software
688 allows for the adjustment of the ROIs to account for peak broadening and slight displacement. A
689 review of the FWHM of the alpha peaks, as calculated by the software, will also reveal peak
690 broadening due to matrix effects and poor test-source preparation.

691 17.3.2.1 Radiochemical Yield

692 Alpha spectrometry test sources are usually prepared by radiochemical separation and the
693 chemical recovery may be less than 100%. Therefore, a radiochemical tracer, which is an isotope
694 of the radioactive species for which the analysis is being performed, may be added to the sample
695 prior to preparation and radioanalysis. The tracer is normally a certified standard solution whose
696 recovered activity is determined during the alpha spectrometric analysis in the same manner as
697 the activities of the isotopes for which the analysis is being performed. The radiochemical yield
698 is then calculated by the spectral analysis program according to:

$$Y = \frac{A_R}{A_S} \quad (17.18)$$

699 where:

700 Y = radiochemical yield

701 A_R = calculated activity recovered

702 A_S = certified activity added (decay corrected to time of counting)

703 The calculation of the chemical yield is normally performed by the alpha spectrometry analysis
704 software using operator input information relative to the alpha energy and abundance, activity,
705 uncertainty, and date of certification of the radiochemical tracer.

706 For some types of radionuclide analyses, no suitable alpha-emitting radionuclide may be
707 available for use as a chemical yield tracer. In this case, the chemical yield may be determined by
708 some other method, such as beta counting, and the resulting yield value provided to the alpha
709 analysis program so the source activity may be calculated from the alpha spectrometry data.

710 When a reference material is used for the chemical tracer, the effective efficiency is measured for
 711 each test source. If the chemical yield is to be reported, an independent measure of the counting
 712 efficiency should be made.

713 17.3.2.2 Uncertainty Calculation

714 The calculation of the combined standard uncertainty for alpha spectrometry is similar to that for
 715 gamma-ray spectrometry as reported in Section 17.3.1.8 above. One additional source of
 716 uncertainty which should be taken into account for alpha spectrometry is that associated with the
 717 determination of radiochemical yield. Since a tracer is added to the sample and the yield
 718 determined by a counting process, the uncertainty involved in this analysis should be accounted
 719 for in the total uncertainty. The uncertainty of the yield determination involves that associated
 720 with the net count of the tracer, the counting efficiency, and that of the emission rate of the tracer
 721 material. The combined standard uncertainty of the radionuclide concentration, R_{C_i} , is given by
 722 either

$$u_c(R_{C_i}) = \sqrt{\frac{u^2(C_{R_i})}{\epsilon_e^2 V^2 e^{-2\lambda_i t_1}} + R_{C_i}^2 \left(\frac{u^2(V)}{V^2} + \frac{u^2(\epsilon_e)}{\epsilon_e^2} \right)} \quad (17.19)$$

723 or

$$u_c(R_{C_i}) = \sqrt{\frac{u^2(C_{R_i})}{\epsilon^2 Y^2 V^2 e^{-2\lambda_i t_1}} + R_{C_i}^2 \left(\frac{u^2(V)}{V^2} + \frac{u^2(\epsilon)}{\epsilon^2} + \frac{u^2(Y)}{Y^2} + \frac{2u(\epsilon, Y)}{\epsilon \cdot Y} \right)} \quad (17.20)$$

724 where:

- 725 C_{R_i} = net count rate in the designated ROI for the radionuclide (cps)
- 726 ϵ = the alpha counting efficiency
- 727 Y = the chemical yield
- 728 ϵ_e = effective efficiency ($\epsilon \cdot Y$) for the tracer (cps/dps)
- 729 V = volume or mass of sample analyzed (L or g)
- 730 e = base of natural logarithm
- 731 λ_i = the radioactive decay constant for the radionuclide (s^{-1} , min^{-1} , or d^{-1})
- 732 t_1 = time lapse from sample collection to beginning of source count (units consistent
 733 with λ_i)
- 734 $u(\cdot)$ denotes the standard uncertainty of a quantity
- 735 $u(\cdot, \cdot)$ denotes the covariance of two quantities

736 The two uncertainty equations are equivalent. However, when the yield is determined using an
737 alpha-emitting tracer, Equation 17.19 generally is easier to implement.

738 **17.3.3 Liquid Scintillation Spectrometry**

739 17.3.3.1 Overview of Liquid Scintillation Counting

740 All modern counters are computer controlled for data acquisition, spectral unfolding, data
741 reduction, sample changer control, external quench correction, and performing the multifarious
742 other functions associated with liquid scintillation counting.

743 Liquid scintillation has traditionally found its primary use in the analysis of low-energy beta
744 emitters, such as ^3H and ^{14}C . In spite of the complicating factors of high background and
745 quenching (Chapter 15), procedures for other beta- and alpha-emitting isotopes have been
746 developed over the years (Holm, 1984; Harvey, 1970).

747 Liquid scintillation has also been applied to the simultaneous analysis of alpha and beta emitters
748 in environmental media (Leyba, 1992). Discrimination between alpha and beta radiation is based
749 on differences in the fluorescence decay pulses. Pulse height is proportional to particle energy,
750 and high counting efficiency results from 4π (4-pi) geometry and the absence of test-source self-
751 attenuation (McDowell and McDowell, 1993). Because of these characteristics, liquid
752 scintillation counting can be utilized as an alternative to gas proportional counting (Section 17.4)
753 and alpha semiconductor counting (Section 17.3.2).

754 17.3.3.2 Liquid Scintillation Spectra

755 The amount of light produced by alpha and beta particles in a liquid scintillation cocktail is
756 proportional to the particle energy. Beta spectra convey the energy continuum from zero to their
757 maximum energy. Alpha liquid scintillation spectra are similar in shape to those obtained by
758 semiconductor spectroscopy, but with greatly decreased resolution. Because alpha particles are
759 only about one-tenth as efficient as beta particles in producing scintillation light pulses, there is
760 an overlap of alpha and beta spectra (Passo and Kessler, 1992; McDowell and McDowell, 1993).

761 Gamma radiation interactions within the scintillation cocktail depend on energy and path length,
762 with lower energy gamma rays being more efficient in transferring their energy. Gamma events
763 are recorded in the same energy range as alpha and beta particles; therefore, discrimination
764 between alpha, beta, and gamma radiation based solely on scintillation spectra is not possible
765 (Passo and Kessler 1992; McDowell and McDowell, 1993).

766 17.3.3.3 Pulse Characteristics

767 Excited triplet and singlet energy states are formed by the fluor molecules when ionizing
768 radiation interacts with the scintillation cocktail. The excited singlet states dissipate their energy
769 very rapidly and produce short lifetime decay pulses, whereas triplet states lose their energy more
770 slowly, resulting in longer lifetime pulses. Because alpha particles have a higher linear energy
771 transfer than gamma or beta radiation, they produce a higher ratio of triplet to singlet excitation
772 states and therefore have a longer pulse duration. Differences in the decay time and shape of the
773 decay pulse are the basis for discriminating of alpha particles from beta and gamma radiation in
774 liquid scintillation counting (Passo and Kessler 1992; Passo and Cook 1994).

775 17.3.3.4 Coincidence Circuitry

776 Most modern liquid scintillation counters employ two photomultiplier tubes 180 degrees apart
777 for the detection of pulses. The light produced when ionizing radiation in the test source interacts
778 with the scintillation cocktail is emitted in all directions. A sample event should therefore
779 produce electronic pulses in both photomultiplier tubes simultaneously, or in coincidence.

780 Electronic noise pulses are produced randomly by the photomultiplier tubes, but the probability
781 that both tubes will produce noise pulses simultaneously is very low. An electronic gate can be
782 set to allow only pulses that are in coincidence to be registered. The rejection of random pulses
783 keeps background counts produced by electronic noise to a minimum.

784 17.3.3.5 Quenching

785 Chemical quenching reduces the amount of energy transferred to the fluor molecules. Halogens,
786 water, solvents, and oxygen are common agents that cause a decrease in the counting efficiency.

787 Color quenching is caused by impurities not removed during test-source preparation or by carrier
788 compounds such as iron chloride. Photons emitted from the fluor molecules are absorbed,
789 reducing the amount of light reaching the photomultiplier tubes.

790 Quenching causes a shift in the scintillation spectrum to lower energies and a reduction in the
791 number of counts. Quenching has a minimal impact on alpha counting, but significantly increases
792 as the energy of the beta particle decreases.

793 The most common method for monitoring quench is through the analysis of the Compton
794 spectrum. After the test source is loaded into the counter, it is irradiated by an external gamma

795 emitting source located in the instrument. The test-source spectrum is collected and compared
796 with factory or user-generated quench standards stored in the instrument library. Both color and
797 chemical quenching cause a shift to lower energies, but the color quench broadens the spectrum
798 as well. The efficiency of the test source is extrapolated and applied to normalize the test-source
799 count rate.

800 17.3.3.6 Luminescence

801 Photoluminescence is produced by ultraviolet light from the environment reacting with the
802 scintillation cocktail. The effect can be minimized by dark adapting the test sources prior to
803 counting.

804 Chemiluminescence is produced by reactions between the scintillation cocktail and chemicals
805 introduced from the test-source preparation. To minimize this effect, oxidizers and alkaline
806 conditions should be avoided.

807 Both photoluminescence and chemiluminescence cause random scintillation events. At low
808 levels, the coincidence gate should reject most of their contribution. However, at very high
809 levels, the probability increases that two events may pass through the gate. Manufacturers use a
810 method of spectral stripping to correct for the false counts, but it is best to avoid the conditions
811 that create the problem.

812 17.3.3.7 Test Source Vials

813 Glass test-source vials contain naturally occurring impurities such as potassium-40, thorium, and
814 uranium. Their contribution appears at the lower energy portion of the spectrum. Plastic vials
815 have a lower background, but they should be compatible with the liquid scintillation cocktail
816 being used. Teflon vials are also available from most manufacturers.

817 17.3.3.8 Data Reduction for Liquid Scintillation Counting

818 Liquid scintillation counters normally provide minimal data reduction in their output. Basic data
819 include the counting duration, count rate in one or more selected windows, and the date and time
820 of counting initiation. A blank source (background) is normally counted with each counting batch
821 and the output will provide the count rate of the blank to be subtracted from each test source.

822 The counting efficiency will also be provided by the output information. Its form of presentation
823 in the output will depend on the calibration/counting (quench correction) method for determining

824 counter efficiency⁴. If the internal (standards addition) method is used the data generated by the
825 counter must be further manipulated in order to develop the counting efficiencies for each test
826 source. When using the external-standards method (quench curve), the scintillation spectrometer
827 will apply the quench corrected efficiency and give the test sample disintegration rate by applying
828 the corrected efficiency.

829 The radionuclide or gross concentration is provided by the following equation:

$$A_C = \frac{C_G - C_B}{\epsilon_q V} \quad (17.21)$$

830 where:

- 831 C_G = the gross counting rate (source + background) (cps)
832 C_B = the counting rate of the blank (cps)
833 ϵ_q = the radionuclide quench corrected counting efficiency (c/d)
834 A_C = radionuclide or gross concentration (Bq/L or Bq/kg)
835 V = the volume or mass analyzed (L or kg)

836 **17.4 Data Reduction on Non-Spectrometry Systems**

837 Proportional counters are primarily used for counting of test sources for alpha and beta emitters.
838 Proportional counters may have entry windows for allowance of the emitted radiation into the
839 active portion of the detector or they may be windowless. These instruments are described in
840 Chapter 15. They are used for the determination of specific radionuclides, following chemical
841 separation to isolate the radionuclide, and for nonspecific (gross) analyses (Chapter 16). Counters
842 are equipped to count alpha and beta simultaneously in a given source and report the activity of
843 both.

844 The basic information obtained from a determination in a proportional counter is the number of
845 counts recorded in the detector within the allotted counting duration. However, modern
846 proportional counters take the data reduction process to the point of finality, i.e., producing the
847 test-source concentration and associated counting uncertainty, providing automatic instrument
848 background subtraction, and correcting for source self-absorption and alpha/beta crosstalk.

⁴ For a discussion of liquid scintillation efficiency determination, see MARLAP Chapter 16, Section 16.5.2.1.

849 The instruments may also have protocols for developing the correction factors for self-absorption
850 and for crosstalk. In addition, they should have the capacity to track and evaluate the periodic
851 quality control checks (check source and background) performed on the instrument.

852 The basic equation used to calculate test-source concentrations is:

$$A = \frac{C_G - C_B}{\epsilon} \quad (17.22)$$

853 where:

854 A = the activity of the radionuclide or gross activity (Bq)
855 C_G = the gross counting rate (source + background) (cps)
856 C_B = the instrument background counting rate (cps)
857 ϵ = the gross or radionuclide counting efficiency (c/d)

858 And the radionuclide or gross concentration is provided by the following equation:

$$A_C = \frac{C_G - C_B}{\epsilon V} \quad (17.23)$$

859 where:

860 A_C = radionuclide or gross concentration (Bq/L or Bq/kg)
861 V = the volume or mass analyzed (L or kg)

862 The associated combined standard uncertainty is given by:

$$u_c(A_C) = \sqrt{\frac{u^2(C_G) + u^2(C_B)}{\epsilon^2 V^2} + A_C^2 \left(\frac{u^2(\epsilon)}{\epsilon^2} + \frac{u^2(V)}{V^2} \right)} \quad (17.24)$$

863 The above simple equations apply to counting either pure alpha or beta emitters and when no
864 correction for self-absorption is necessary (weightless sources). Modifications should be made in
865 the activity and concentration calculations when both alpha and beta particles are emitted by the
866 source, and when absorption and scattering within the source cause a reduction in the effective
867 efficiency.

868 Self-absorption factors are applied for sources where the internal attenuation of the alpha or beta
 869 particle is sufficient to affect the overall efficiency (Chapter 16). Commercially available
 870 proportional counters have a protocol for developing the self-absorption correction factors. These
 871 protocols process the data generated by counting a series of alpha calibration sources and a series
 872 of beta calibration sources, which both have varying masses of material, from “zero” to the
 873 maximum to be encountered in test sources (Chapter 16). The instrument is programmed to then
 874 fit the data to a mathematical function so the counting efficiency correction factor can be applied
 875 at any test-source mass within the range covered by the calibration source masses. A cubic
 876 polynomial is one option used for both alpha and beta counting efficiencies. A cubic polynomial
 877 has the form

$$\epsilon_m = a_0 + a_1 m + a_2 m^2 + a_3 m^3 \quad (17.25)$$

878 where:

- 879 m = is the residual mass of the test source
 880 ϵ_m = the counting efficiency at mass m
 881 a_i = constants determined by the data fit

882 The combined standard uncertainty of ϵ_m is given by

$$u_c(\epsilon_m) = \sqrt{u^2(a_0) + \sum_{i=1}^3 m^{2i} u^2(a_i) + 2 \sum_{i=0}^2 \sum_{j=i+1}^3 m^{i+j} u(a_i, a_j) + (a_1 + 2a_2 m + 3a_3 m^2)^2 u^2(m)} \quad (17.26)$$

883 When the identities of the alpha or beta emitting radionuclides are unknown, an additional
 884 component of uncertainty is needed to account for the dependence of the counting efficiency (and
 885 self-absorption) on the unknown particle energy.

886 Another option that is often used for the beta counting efficiency is an exponential curve, which
 887 has the form

$$\epsilon_m = \epsilon_{\text{zero}} e^{-am} \quad (17.27)$$

888 where:

- 889 m = is the residual mass of the test source
 890 ϵ_m = the counting efficiency at mass m
 891 ϵ_{zero} = the “zero” mass counting efficiency
 892 a = constant determined by the data fit

893 Then the combined standard uncertainty of ϵ_m is:

$$u_c(\epsilon_m) = e^{-am} \sqrt{a^2 u^2(m) + u^2(\epsilon_{\text{zero}}) + m^2 u^2(a) - 2m u(\epsilon_{\text{zero}}, a)} \quad (17.28)$$

894 Again, an additional uncertainty component may be needed when the identity of the beta-emitting
895 radionuclide is unknown.

896 Crosstalk, sometimes called “spill over,” refers to the misclassification of alpha- and beta-
897 produced counts in a proportional counter which is designed to count both particles
898 simultaneously. It occurs when counts produced by alpha interactions in the detector are
899 registered as beta counts and vice versa. In order to accurately record the alpha and beta activities
900 of sources containing radionuclides emitting both particles, corrections must should be made for
901 crosstalk.

902 The number of alpha interactions registered as beta counts will increase as the source self-
903 absorption increases. The opposite is true for beta crosstalk, in that the number of beta
904 interactions falsely designated as alpha counts decreases with source self-absorption. Thus,
905 crosstalk correction factors vary with test-source mass and should be developed for the range of
906 test-source masses to be encountered. Commercially available proportional counters have
907 established programs to assist in the establishment of alpha and beta crosstalk factors. The
908 algorithms to correct for crosstalk are presented below.

909 The alpha in beta crosstalk, X_α , is defined as:

$$X_\alpha = \frac{\beta}{\alpha + \beta} \quad (17.29)$$

910 The respective counts in the alpha channel (α) and those in the beta channel (β) counts are
911 measured with a pure alpha-emitting source. Likewise, the beta in alpha crosstalk, X_β , is:

$$X_\beta = \frac{\alpha}{\alpha + \beta} \quad (17.30)$$

912 The respective alpha (α) and beta (β) count rates are measured with a pure beta-emitting source.

913 The relationship between X_α and X_β is given by:

$$\alpha = \alpha_d - \alpha_d X_\alpha + \beta_d X_\beta \quad (17.31)$$

$$\beta = \beta_d - \beta_d X_\beta + \alpha_d X_\alpha \quad (17.32)$$

914 Equation 17.31 states that the recorded alpha count rate, α , consists of the actual alpha count rate,
 915 α_d , (the total alpha count rate in both the alpha and beta channels due to only alpha interactions),
 916 minus those alpha interactions recorded in the beta channel, plus those beta counts recorded in
 917 the alpha channel. Equation 17.32 states the equivalent of Equation 17.31 for beta counts.
 918 Solving the equations simultaneously for α_d and β_d gives:

$$\alpha_d = \frac{\alpha - X_\beta (\alpha + \beta)}{1 - X_\alpha - X_\beta} \quad (17.33)$$

$$\beta_d = \frac{\beta - X_\alpha (\alpha + \beta)}{1 - X_\alpha - X_\beta} \quad (17.34)$$

919 Their associated combined standard uncertainties are:

$$u_c(\alpha_d) = \frac{\sqrt{u^2(X_\alpha)\alpha_d^2 + u^2(X_\beta)(\alpha_d - \alpha - \beta)^2 + u^2(\alpha)(1 - X_\beta)^2 + u^2(\beta)X_\beta^2}}{1 - X_\alpha - X_\beta} \quad (17.35)$$

$$u_c(\beta_d) = \frac{\sqrt{u^2(X_\beta)\beta_d^2 + u^2(X_\alpha)(\beta_d - \alpha - \beta)^2 + u^2(\beta)(1 - X_\alpha)^2 + u^2(\alpha)X_\alpha^2}}{1 - X_\alpha - X_\beta} \quad (17.36)$$

920 Since crosstalk factors vary with radionuclide, additional uncertainty components may be needed
 921 when the identities of the alpha and beta emitting radionuclides are unknown.

922 Processors execute many other functions for instruments which do not perform spectrometry.
 923 These instruments include proportional counters, scintillation detectors, ionization chambers, and
 924 special instruments (Chapter 15). The functions performed by processors may include instrument
 925 control (sample change, gas flow control, etc.) and the calculations necessary to convert the basic
 926 counting information to final form data or to some intermediate step.

927 Data reduction functions which may be performed for scintillation detectors, ionization
928 chambers, and special instruments include the following:

- 929 • Background determination and subtraction;
- 930 • Conversion of total counts to counts per second;
- 931 • Calculate activity using calibration data;
- 932 • Calculate concentration using activity and operator input data;
- 933 • Perform efficiency calibrations;
- 934 • Calculate counting and total uncertainty;
- 935 • Cross talk determination and correction;
- 936 • Self-absorption corrections;
- 937 • Radioactive decay corrections; and
- 938 • Quality control (QC) functions (efficiency and background verification).

939 The output of manual systems usually requires further reduction to render it usable. The
940 information generated by processor-based systems may also need further processing.

941 These additional calculations may be performed using a calculator or by a computer using
942 general or custom software programs. The data may be electronically transferred to the
943 processing computer by a local area network (LAN) or on a computer disk. In some cases the
944 processing software may be part of the LIMS.

945 **17.5 Reporting Data**

946 Quality assurance planning documents will give the level of data reporting required. This level
947 will vary from simply confirming the presence or absence of an analyte to a complete reporting
948 of all measurements, calibration data, documentation of the performance of laboratory processes,
949 provision of certain instrument counting reports, and QC sample results and analysis. Another
950 way of viewing this is as a tiered approach where preliminary studies or site surveys may only
951 require a minimum of data reporting, while a final site survey may require a detailed reporting of
952 the results. The necessary elements for data reporting are connected to the purpose for which the
953 data will be used (data quality objectives).

954 MARLAP recommendations for data reporting are that the reported value of a measurement
955 result: (1) be reported directly as obtained, with appropriate units, even if they are negative
956 values, (2) be expressed in an appropriate number of significant figures, and (3) include an
957 unambiguous statement of the uncertainty. The appropriate number of significant figures is
958 determined by the magnitude of uncertainty in the reported value. Each reported measurement

959 result should include the value and an estimate of the uncertainty (expanded uncertainty) (ANSI
960 42.23).

961 **17.5.1 Sample and Analysis Method Identification**

962 Sample data are normally reported by sample number, including both the field (project) and
963 laboratory assigned identifiers. In addition, the submitting laboratory should be identified as well
964 as the analysis method (ANSI 42.23, p. 38). Other information which can assist in the review and
965 interpretation of the data may be requested. This could include sample collection date (decay
966 correction reference date), analysis date, chain-of-custody (COC) number, and site or project
967 name.

968 **17.5.2 Units and Radionuclide Identification**

969 The individual radionuclides should be identified or, for gross analyses, the category, e.g., gross
970 alpha/beta, should be reported. Reporting units are likely specified by project planning
971 documents. If not specified, when possible, International System of Units (SI) units are preferred.
972 However, since regulatory compliance levels are usually quoted in traditional radiation units, it
973 may be appropriate to report in both SI and traditional units with one being placed within a
974 parenthesis. Both the SI and non-SI units are shown in Table 17.1 for common matrices.

TABLE — 17.1 Units For Data Reporting

Matrix	In Non-SI Units	In SI Units	Conversion Factor From Non-SI to SI Units
Airborne Particulates and Gas	pCi m ⁻³	Bq m ⁻³	3.70 × 10 ⁻²
Liquids	pCi L ⁻¹	Bq L ⁻¹	3.70 × 10 ⁻²
Solids	pCi kg ⁻¹ or pCi g ⁻¹	Bq kg ⁻¹	3.70 × 10 ⁻² or 37
Surfaces	dpm / 100 cm ²	Bq / 100 cm ²	1.67 × 10 ⁻²

975 **17.5.3 Values, Uncertainty, and Significant Figures**

976 The value, as measured, including zero and negative numbers, and the measurement uncertainty
977 (either expanded uncertainty or the combined standard uncertainty) should be reported in the
978 same units (Chapter 19). In general, environmental radiation measurements seldom warrant more
979 than two or three significant figures for the reported value, and one or two significant figures for
980 the uncertainty. As recommended in Chapter 19, Section 19.3.6, the measurement uncertainty
981 should be rounded to two significant figures, and both the value and uncertainty reported to the

982 resulting number of decimal places. For example, a value of 0.8961 pCi/L with an associated
983 measurement uncertainty of 0.0234 should be reported as 0.896 ± 0.023 pCi/L. The MDC should
984 be reported to two significant figures (ANSI 42.23, p38). It should be noted that truncation
985 should only occur in reporting the final results (Section 18.3.6).

986 **17.5.4 Other Information to be Provided on Request**

987 Information which should be documented and retained for provision, if requested, includes
988 (ANSI 42.23, p38):

- 989 • Total weight or volume of the sample submitted and analyzed;
- 990 • Identification and documentation of specific analysis processes and analyst;
- 991 • Specific analytical parameters, i.e., chemical yields, counting times, decay factors, efficiency
992 of detectors used;
- 993 • Date, time, and place of sampling;
- 994 • Sample receipt information; and
- 995 • QC data demonstrating the quality of the measurement.

996 **17.6 Data Packages**

997 Project planning documents (Chapter 4) and analytical statements of work (Chapter 5) will
998 usually define the requirements of the final data submittal. Many projects will specify a data
999 package which contains not only the data reports described in the preceding section, but other
1000 supporting information to further describe, document, and define the analytical process. These
1001 additional requirements may be instituted to provide a basis for data verification/validation
1002 (Chapter 8), the purpose of which is to confirm that the data meet project quality objectives
1003 (Chapter 2). Material which may be required as part of a data package is discussed in Chapter 5.

1004 **17.7 Electronic Data Deliverables**

1005 Many project planning documents and SOWs require that laboratory data be delivered in
1006 electronic format, commonly called electronic data deliverables (EDD). This allows the data to
1007 be directly entered into a project database or, in some cases, into validation programs, and avoids

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1008 transcription errors. There is no universal format for presenting EDDs, so the laboratory may be
1009 required to produce them in various formats. While the record structure of the EDD may vary in
1010 terms of the length and order of the fields, it is likely that the following are examples that may be
1011 requested:

- 1012 • Field Sample Number
- 1013 • Laboratory Sample Number
- 1014 • Sample Collection or Reference Date
- 1015 • Sample Receipt Date
- 1016 • Analysis Date
- 1017 • Result Identifier (sample or type of QC sample)
- 1018 • Radionuclide
- 1019 • Result
- 1020 • Results Units
- 1021 • Measurement Uncertainty
- 1022 • Sample Aliquant Size
- 1023 • Aliquant Size Units
- 1024 • Minimum Detectable Concentration
- 1025 • Minimum Quantifiable Concentration (MQC)

1026
1027 More information on EDDs may be found at the following websites:

1028 More information on EDDs may be found at the websites listed here. The U.S. Department of
1029 Energy EDD may be found at: (<http://www.em.doe.gov/namp/pitimp.html>) or (<http://www.em.doe.gov/namp/deemmeet.html>). Another EDD that is more general has been developed. It
1030 is called the General Electronic Data Deliverable (GEDD) and may be found at the website:
1031 (<http://ersmo.inel.gov/edd/gedd.html#Entity Relationship Diagram>). The EPA Environmental
1032 Data Registry may be found at: (<http://www.epa.gov/edr/>). U.S. Air Force Environmental
1033 Resources Program Management System (ERPRIMS) website: (http://www.afcee.brooks.af.mil/ms/msc_irp.htm) also provides useful information on environmental databases and
1034 EDDs.
1035
1036

1037 EDDs may be transmitted by direct electronic transfer, e-mail, or by diskette.

1038 **17.8 References**

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