

SECTION 1
GROSS ALPHA AND GROSS BETA RADIOACTIVITY IN DRINKING WATER
METHOD 900.0

1.0 Scope and Application

- 1.1** This method covers the measurement of gross alpha and gross beta particle activities in drinking water. The method is a screening technique for monitoring drinking water supplies for alpha and beta particle activities according to the limits set forth under the Safe Drinking Water Act, PL 93-523, 40 FR 34324, and thereby determining the necessity for further analysis.
- 1.2** The method is applicable to the measurement of alpha emitters having energies above 3.9 megaelectronvolts (MeV) and beta emitters having maximum energies above 0.1 MeV.
- 1.3** The minimum limit of concentration to which this method is applicable depends on sample size, counting system characteristics, background, and counting time. The National Primary Interim Drinking Water Regulations (NIPDWR) require a gross beta detection limit of 4 pCi/L, an alpha detection limit of 1 pCi/L for compliance with Part 141.15(a) and a gross alpha detection limit of 3 pCi/L for compliance with Part 141.15(b).
- 1.4** Since, in this method for gross alpha and gross beta measurement, the radioactivity of the sample is not separated from the solids of the sample, the solids concentration is very much a limiting factor in the sensitivity of the method for any given water sample. Also, for samples with very low concentrations of radioactivity such as from drinking water sources, it is essential to analyze as large a sample aliquot as is needed to give reasonable counting times in meeting the required sensitivities (detection limits) indicated above. The Regulations define sensitivity in terms of detection limits Part 141.25(c) of the Regulations.
- 1.5** The largest sample aliquot that should be counted for gross alpha activity is that size aliquot which gives a solids density thickness of 5 mg/cm² in the counting planchet. For a 2-inch diameter counting planchet (20 cm²), an aliquot containing 100 mg of dissolved solids would be the maximum aliquot size for that sample which should be evaporated and counted for gross alpha activity.
- 1.6** When the concentration of total dissolved solids (TDS) is known for a given water sample and the alpha background and the counting efficiency of a given counting system are known, the counting time that is needed to meet the required sensitivity (3 pCi/L) can be determined by equations given in Appendix C.
- 1.7** For the counting of gross beta activity in a water sample the TDS is not as limiting as for gross alpha activity because beta particles are not stopped in solids as easily as are alpha particles. Very often a single sample aliquot is evaporated and counted for both gross alpha and gross beta activity. In that case the sample aliquot size would be dictated by the solids limitations for alpha particles. For water samples that are to be counted for gross beta activity, equations in Appendix C can also be used to

determine the necessary counting time to meet a sensitivity for gross beta activity (4 pCi/L required by NIPDWR).

1.8 Radionuclides that are volatile under the sample preparation conditions of this method will not be measured. In some areas of the country the nitrated water solids (sample evaporated with nitric acid present) will not remain at a constant weight after being dried at 105°C for two hours and then exposed to the atmosphere before and during counting. Other radioactivities may also be lost during the sample evaporation and drying at 105°C (such as some chemical forms of radioiodine). Those types of water samples need to be heated to a dull red heat for a few minutes to convert the salts to oxides. Sample weights are then usually sufficiently stable to give consistent counting rates and a correct counting efficiency can then be assigned. Some radioactivities, such as the cesium radioisotopes, may be lost when samples are heated to dull red color. Such losses are limitations of the test method.

1.9 This method provides a rapid screening measurement to indicate whether specific analyses are required. For drinking waters with an extremely high solids content (>500 ppm), method 900.1 is recommended.

2.0 Summary of Method

2.1 An aliquot of a preserved drinking water sample is evaporated to a small volume and transferred quantitatively to a tared 2-inch stainless steel counting planchet. The sample residue is dried to constant weight, reweighed to determine dry residue weight, then counted for alpha and/or beta radioactivity.

2.2 Counting efficiencies for both alpha and beta particle activities are selected according to the amount of sample solids from counting efficiency vs sample solids standard curves.

3.0 Sample Handling and Preservation

3.1 A representative sample must be collected from a free-flowing source of drinking water, and should be large enough so that adequate aliquots can be taken to obtain the required sensitivity.

3.2 It is recommended that samples be preserved at the time of collection by adding enough 1N HNO₃ to the sample to bring it to pH 2 (15 mL 1N HNO₃ per liter of sample is usually sufficient.) If samples are to be collected without preservation, they should be brought to the laboratory within 5 days, then preserved and held in the original container for a minimum of 16 hours before analysis or transfer of the sample.

3.3 The container choice should be plastic over glass to prevent loss due to breakage during transportation and handling.

4.0 Interferences

4.1 Moisture absorbed by the sample residue is an interference as it obstructs counting

and self-absorption characteristics. If a sample is counted in an internal proportional counter, static charge on the sample residue can cause erratic counting, thereby preventing an accurate count.

- 4.2 Non-uniformity of the sample residue in counting planchet interferes with the accuracy and precision of the method.
- 4.3 Sample density on the planchet area should not be not more than 5 mg/cm² for gross alpha and not more than 10 mg/cm² for gross beta.
- 4.4 When counting alpha and beta particle activity by a gas flow proportional counting system, counting at the alpha plateau discriminates against beta particle activity, whereas counting at the beta plateau is sensitive to alpha particle activity-present in the sample. This latter effect should be determined and compensated for during the calibration of the specific instrument being used.

5.0 Apparatus - See Appendix D for details and specifications.

- 5.1 Gas-flow proportional counting system, or
- 5.2 Scintillation detector system
- 5.3 Stainless steel counting planchets
- 5.4 Electric hot plate
- 5.5 Drying oven
- 5.6 Drying lamp
- 5.7 Glass desiccator
- 5.8 Glassware
- 5.9 Analytical balance

6.0 Reagents

All chemicals should be of "reagent-grade" or equivalent whenever they are commercially available.

- 6.1 Distilled or deionized water having a resistance value between 0.5 and 2.0 megohms (2.0 to 0.5 micromhos)/cm at 25°C.
- 6.2 Nitric acid, 1N: Mix 6.2 mL 16N HNO₃ (conc.) with deionized or distilled water and dilute to 100 mL.

7.0 Calibrations

- 7.1** For absolute gross alpha and gross beta measurement, the detectors must be calibrated to obtain the ratio of count rate to disintegration rate. Americium-241 (used for alpha activity in the collaborative test of this method) has higher alpha particle energy (5.49 MeV) than those emitted by the naturally occurring uranium and radium-226 radionuclides but is close to the energy of the alpha particles emitted by naturally occurring thorium-228 and radium-224. Standards should be prepared in the geometry and weight ranges to be encountered in these gross analyses. It is, therefore, the prescribed radionuclide for gross alpha calibration. NBS or NBS-traceable americium-241 is available from Standard Reference Materials Catalog, NBS Special Publications 260, U.S. Department of Commerce (1976) and from Quality Assurance Branch, EMSL-LV, P.O. Box 15027, Las Vegas, Nevada 89114.
- 7.2** Strontium-90 and cesium-137 have both been used quite extensively as standards for gross beta activity. Standard solutions of each of these radionuclides are readily available. Cesium is volatile at elevated temperatures (above 450°C). Some water supplies have dissolved solids (salts) that, when converted to nitrate salts, are quite hygroscopic and need to be converted to oxides by heating to red heat to obtain sample aliquots that are weight-stable. Sample weight stability is essential to gross alpha and gross beta measurements to ensure the accuracy of the self-absorption counting efficiency factor to be used for the samples. Strontium-90 in equilibrium with its daughter yttrium-90 is the prescribed radionuclide for gross beta calibrations.
- 7.3** For each counting instrument to be used, the analyst should prepare separate alpha and beta particle self-absorption graphs showing water sample residue weight (mg) vs the efficiency factor (dpm/cpm), using standard alpha and beta emitter solutions and tap water. For the alpha graph standard, alpha activity is added to varying size aliquots of tap water, such that the aliquot residue weight is varied between 0 and 100 mg (for a 2-inch counting planchet). A similar graph is prepared with standard beta activity and tap water aliquots, varying the residue weight between 0 and 300 mg (for a 2-inch planchet). If it is planned to use water sample aliquot volumes that always contain 100 mg of dried water solids, then only the efficiency factor for that residue weight needs to be established.
- 7.4** Tap water aliquots with added americium-241 or strontium-90 standard should be acidified with a few mL 16N HNO₃, evaporated to a small volume in a beaker on a hot plate, transferred quantitatively in 5 mL portions or less to a tared counting planchet, evaporated to dryness, and finally dried at 105°C for 2 hours (or flamed to a red heat if dried solids appear to be noticeably hygroscopic). Weight-stable aliquot residues should then be alpha and/or beta counted until at least 10,000 total counts have been accumulated. A single set of reference standards prepared in this way can be used for each counting instrument for separate graph preparations and can be stored for reverification whenever needed.

8.0 Procedure

- 8.1** Transfer to a beaker an aliquot of a water sample of a volume size that contains no more than 100 mg (for alpha only or alpha and beta determination) or 200 mg (for beta only determination) of total water solids. Evaporate the aliquot to near dryness on a hot plate. If water samples are known or suspected to contain chloride salts,

those chloride salts should be converted to nitrate salts before the sample residue is transferred to a stainless steel planchet (Chlorides will attack stainless steel and increase the sample solids and no correction can be made for those added solids). Chloride salts can be converted to nitrate salts by adding 5 mL portions of 16N HNO₃ to the sample residue and evaporating to near dryness. (Two treatments are usually sufficient.) Add 10 mL 1N HNO₃ to the beaker and swirl to dissolve the residue. Quantitatively transfer the aliquot concentrate in small portions (not more than 5 mL at a time) to a tared planchet, evaporating each portion to dryness.

- 8.2** Dry the sample residue in a drying oven at 105°C for at least 2 hours; cool in a desiccator; weigh; and count. Store the sample residue in a desiccator until ready for counting.
- 8.3** Some types of water dissolved solids, when converted to nitrate salts, are quite hygroscopic even after being dried at 105°C for two hours. When such hygroscopic salts are present with samples that are put into an automatic counting system, those samples gain weight while they are waiting to be counted and inaccurate counting data result. When there is evidence of hygroscopic salts in sample counting planchets, it is recommended that they be flamed to a dull red heat with a Meeker burner for a few minutes to convert the nitrate salts to oxides before weighing and counting.
- 8.4** Count for alpha and beta activity at their respective voltage plateaus. If the sample is to be recounted for reverification, store it in a desiccator.

Note: As long as counting chambers are capable of handling the same size planchet, alpha and beta activity can be determined at their respective voltage plateaus in the designated counting instruments. Keep planchet in the desiccator until ready to count because vapors from moist residue can damage detector and window and can cause erratic measurements. Samples may be counted for beta activity immediately after drying; but alpha counting should be delayed at least 72 hours until equilibrium has occurred. If the gas-flow internal proportional counter does not discriminate for the higher energy alpha pulses at the beta plateau, the alpha activity must be subtracted from the beta plus alpha activity. This is particularly important for samples with high alpha activity.

9.0 Calculations

- 9.1** Calculate the alpha radioactivity by the following equation:

$$\text{Alpha (pCi/L)} = \frac{A \times 1000}{2.22 \times C \times V}$$

where:

A = net alpha count rate (gross alpha count rate minus the background count rate) at the alpha voltage plateau

C = alpha efficiency factor, read from graph of efficiency versus mg of water solids per cm² of planchet area, (cpm/dpm)

V = volume of sample aliquot, (mL)

2.22 = conversion factor from dpm/pCi

9.2 Calculate the beta radioactivity by the following equations:

9.2.1 If there are no significant alpha counts when the sample is counted at the alpha voltage plateau, the beta activity can be determined from the following equation:

$$\text{Beta (pCi/L)} = \frac{B \times 1000}{2.22 \times D \times V}$$

where :

B = net beta count rate (gross count rate minus the background count rate at the beta voltage plateau)

D = beta efficiency factor, read from the graph of efficiency versus mg of water solids per cm² of planchet area, (cpm/dpm)

V = volume of sample aliquot, (mL)

2.22 = conversion factor from dpm/pCi

9.2.2 When counting beta radioactivity in the presence of alpha radioactivity by gas-flow proportional counting systems (at the beta plateau) alpha particles are also counted. Since alpha particles are more readily absorbed by increasing sample thickness than beta particles, the alpha/beta count ratios vary with increasing sample thickness. Therefore, it is necessary to prepare a calibration curve by counting standards containing americium-241 with increasing thickness of solids on the alpha plateau and then on the beta plateau, plotting the ratios of the two counts vs density thickness. The alpha amplification factor (E) from that curve is used to correct the amplified alpha count on the beta plateau. When significant alpha activity is indicated by the sample count at the alpha voltage plateau, the beta activity of the sample can be determined by counting the sample at the beta voltage plateau and calculating the activity from the following equation:

$$Beta (pCi/L) = \frac{(B - AE) \times 1000}{2.22 \times D \times V}$$

where:

B = (as defined above)

D = (as defined above)

A = (as defined above)

E = alpha amplification factor, read from the graph of the ratio of alpha counted at the beta voltage/alpha counted at the alpha voltage vs sample density thickness

V = volume of sample aliquot, (mL)

2.22 = conversion factor from dpm/pCi.

9.3 Errors associated with the results of the analysis should also be reported. (See Appendix B for error and statistical calculations).

10.0 Precision and Accuracy

10.1 In an interlaboratory collaborative test of the method, three sets of samples were analyzed by 18 laboratories for gross alpha and gross beta activity. The samples were prepared with dissolved water solids with known additions of americium-241 for gross alpha and cesium-137 for gross beta activity. Sample series A contained only americium-241 radioactivity, series B contained only cesium-137 radioactivity, and series C contained both americium-241 and cesium-137 radioactivities. Participating laboratories were supplied with standard solutions of americium-241 and cesium-137 and blank solution of dissolved water solids for preparing sample self-absorption curves.

10.2 The gross alpha data from two laboratories was rejected for the statistical analysis because their scores in the ranked results of the laboratory averages were out of the acceptable range for 18 laboratories. The gross beta data from 3 laboratories were rejected for the statistical analysis for the same reason.

10.3 The coefficients of variation for the combined within-laboratory precision for gross alpha analysis of the 3 samples ranged from 7.4% to 12.2%. The coefficients of variation for the precision of the method between laboratories ranged from 11.5% to 14.6% for gross alpha analysis for the 3 samples.

10.4 The coefficients of variation for the combined within-laboratory precision for gross beta analysis for the 3 samples ranged from 3.5% to 5.2%. The coefficients of variation for the precision between laboratories for gross beta analysis for the 3

samples ranged from 3.5% to 7.5%. The coefficients of variation for the total error between laboratories based on a single analysis ranged from 5.9% to 8.3% for gross beta analysis of the 3 samples.

- 10.5** In the statistical test to detect method bias the calculated values for "t" were well below the specified critical value for "t" for both gross alpha and gross beta analysis, indicating no bias in the method. Also, a comparison of the known values to the grand average values shows a deviation of less than 10% for alpha activity for the 3 samples. The same comparison for beta activity shows a deviation of less than 2% for each of the 3 samples.
- 10.6** Whenever the same radioisotopes are present in standards and samples, acceptable accuracy of measurement of alpha and beta activities would be expected. Whenever different radioisotopes are present in standards and samples, especially when significantly different particle energies are involved, then any measurement of gross alpha and gross beta activity in the sample will only be an estimation of the true activities. Such an estimation can only serve to indicate the need for more specific analyses.

Bibliography

1. Standard Methods for The Examination of Water and Waste Water, 14th Ed., American Public Health Association, Washington, D.C. (1976).
2. 1979 Annual Book of ASTM Standards, Part 31, American Society for Testing and Materials, Philadelphia, Pa. (1979)
3. Friedlander, G., J.W. Kennedy, and J. Miller, Nuclear and Radiochemistry, John Wiley and Sons, Inc., New York, New York, 1964.
4. Youden, W.J., and E.H. Steiner, Statistical Manual of the Association of Official Analytical Chemists, 1975.
5. Dixon, W.J., and F.J. Massey, Jr., Introduction to Statistical Analysis, 3rd Edition, McGraw-Hill, 1969.
6. Harley, J.H., N.A. Hallden, and I.M. Fisenne, "Beta Scintillation Counting With Thin Plastic Phosphors," Nucleonics, 20:59, 1961.
7. Hallden, N.A.. and J.H. Harley, "An Improved Alpha-Counting Technique," Analytical Chemistry, 32:1861, 1960.