USTUR 600: ALPHA SPECTROMETRY MEASUREMENT FOR RADIONUCLIDES OF AMERICIUM, PLUTONIUM, URANIUM, AND THORIUM

Purpose	Alpha spectrometry measurement for radionuclides of americium, plutonium, uranium, and thorium	**Method Number**	USTUR600
Original Date	10/10/95	**Author**	USTUR Radiochemistry Staff
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1. Principle of Method

1.1. Samples which have been mounted according to USTUR 500 or 510 are measured for alpha activity using alpha spectrometry.

1.2. Detection limits for alpha activity are based on instrument background count data collected in region of interest areas associated with the alpha decay spectrum for each nuclide of interest, the recovery of the radiotracer, the counter efficiency, the branching ratio of the nuclide of interest, the length of the sample count, and the background time.

1.3. Multinuclide alpha secondary standards are used to energy- and efficiency-calibrate the alpha spectrometers according to USTUR 620.

1.4. Quality control methods are used to monitor counter efficiency and isotopic backgrounds to insure detectors remain in control. (See USTUR 620).

1.5. See USTUR 660 for samples to be measured by absolute analysis.

1.6. See USTUR 650 for samples to be measured by relative analysis.

2. Apparatus

2.1. Alpha spectrometer: 4 unit ORTEC Octete PC, 4096-channel analyzer.

2.2. Vacuum pump.

2.3. 450 mm^2 EG&G ORTEC ULTRA silicon surface barrier detectors.

2.4. Alphavision V4.02.

2.5. Maestro for Windows V.5.0.

2.6. Forceps.

3. Alpha Spectroscopy Overview

3.1. Each alpha spectrometer is set up with an energy range of 3.5 to 6 MeV in 512 channels.

3.2. Each detector is energy-calibrated using multinuclide secondary standards of ^{242}Pu, ^{239}Pu, and ^{241}Am prepared according to USTUR 630. See USTUR 620 for the energy-calibration procedures. Table 1 shows the energy and channels for the calibration nuclides.

Table 1

Isotope	Principal α Energy (keV)	Channel
Pu-242	4901	280
Pu-239	5156	331
Am-241	5486	397

3.3. Each detector is efficiency-calibrated using the ^{242}Pu content of the secondary alpha sources. See USTUR 620 for the efficiency-calibration procedure.

3.4. A 300,000 s background count typically is obtained and a control chart kept for each isotopic region of interest (ROI) for each detector. See USTUR 620 for background counting procedures and QA/QC methods associated with backgrounds.

3.4.1. All detectors to be used for analysis shall have had an acceptable background measurement within four weeks prior to beginning the analysis.

3.4.2. The last background measurement is used for background subtraction as defined in the equations in Section 5.

3.5. Samples are routinely measured for 150,000 s on shelf 2.

3.5.1. Tracers with impurities are discouraged from use for sample analysis. Tracers for which no impurity free source exists are used in some cases (Am-243 contains a small amount of Am-241).

3.5.2. Contamination in tracers is corrected by subtracting the fraction of isotopic activity from the net isotopic count rate as described in Section 5.

Example: The net counts in the ROI for Am-243 tracer was 2000 and the net counts for Am-241 was 100. This tracer is known to be contaminated with 0.17% (counts ^{241}Am/counts ^{243}Am) Am-241. Thus (2000 counts Am-243) x 0.0017 counts ^{241}Am/counts ^{243}Am = 3.4 counts which are subtracted from the net counts for Am-241 resulting in 96.6 counts.

3.6. Chambers for Th and U measurement are equipped with recoil suppression and are operated at a pressure of ~35 torr.

3.7. Blanks are run with each batch to monitor for reagent contribution as well as to provide monitoring for possible contamination during analysis.

3.7.1. QC charts of blank values are evaluated to insure blank measurements are in control.

3.7.2. Blanks are corrected for tracer recovery according to relative analysis procedures described in USTUR 650.

3.7.3. Blanks are corrected for known tracer contamination as described in Section 3.5.

3.7.4. Sample activities are corrected by subtracting the average isotopic blank value if necessary.

3.7.5. Batches with blanks deemed out of control (exceed 99% confidence limits) are evaluated for cause of discrepancy and re-run if possible. If the sample cannot be re-run, the blank value from the batch will be used for blank subtraction. Appropriate indication of the correction method used shall be indicated on the analysis form.

3.8. Samples are evaluated by measuring the activity of isotopes in a specific region of interest (ROI) following radiochemical separation and sample mounting.

3.9. Absolute analysis of a planchet is conducted according to USTUR 660. Absolute analysis is used for limited applications such as secondary source preparation and background counts.

3.10. Relative analysis of a sample is conducted according to USTUR 650. Relative analysis is the standard method used to evaluate USTUR radiochemical samples.

3.11. ROI’s and FWHM (full width at half-maximum)

3.11.1. The alpha emissions of most radionuclides are made up of several different alpha energies each with a specific decay probability. Table 1 shows the emission probabilities of selected radionuclides.

3.11.2. The size of the FWHM will dictate the size of the ROI, the number of peaks observable, and may cause the alpha decays from one isotope to overlap with another of the same element. Table 2 indicates the ROI changes as the FWHM increases. The effective branching ratio of a radionuclide may also change as the ROI expands to include minor alpha peaks.

4. Data Quality

The USTUR program requires high quality data in support of its efforts to provide improved understanding of biokinetic modelling, distribution studies, and other operations. Data quality criteria are provided by USTUR-Richland prior to sample analysis. Figures 2, 3, 4, and 5 may be useful in establishing these objectives.

5. Sample calculations for relative analysis

5.1. Calculation of aliquot activity (A_a)

$$A_a\,(\text{dpm}) = \frac{A_{0t} \cdot B_t\left(c_s t_b - c_b t_s - v_t\left(c_{st} \cdot t_b - c_{bt} \cdot t_s\right)\right)}{B_s\left(c_{st} t_b - c_{bt} t_s\right)}\, e\left[\frac{\ln 2}{t_{t,1/2}}\right]\left[d_c - d_d\right]\frac{1\text{ year}}{365.25\text{ days}}, \text{ where}$$

A_{0t} = Activity of the tracer at date of certification (dpm)
$T_{t,1/2}$ = Tracer half-life (years)
d_d = Date of detection (counting)
d_c = Date of certification (tracer)
c_s = Gross counts of sample
c_b = Background counts of sample
t_b = Background time of sample in seconds
c_{st} = Gross counts of tracer
c_{bt} = Background counts of tracer
B_t = Tracer branching ratio
B_s = Nuclide branching ratio
t_s = Sample count time in seconds
v_t = Relative count rate of contamination of nuclide in tracer.

5.2. Calculation of uncertainty in the activity for the aliquot $\left(\sigma_{A_a}\right)$

$$\sigma_{A_a} = \left| A_a \sqrt{\left(\frac{\sigma_{A_{0t}}}{A_{0t}}\right)^2 + \frac{\left(c_{st} \cdot t_b^2 + c_{bt} \cdot t_s^2\right)\left(c_s \cdot t_b - c_b \cdot t_s\right)^2}{\left(c_{st} t_b - c_{bt} t_s\right)^2\left(c_s t_b - c_b t_s - v_t\left(c_{st} t_b - c_{bt} t_s\right)\right)^2} + \frac{c_s t_b^2 + c_b t_s^2 + \sigma_{v_t}^2\left(c_{st} t_b - c_{bt} t_s\right)^2}{\left(c_s t_b - c_b t_s - v_t\left(c_{st} t_b - c_{bt} t_s\right)\right)^2}} \right|$$

σ_{A0t} = The uncertainty associated with the activity of the tracer at date of certification (dpm).

σ_{Vt} = Uncertainty in relative contamination of nuclide in tracer.

5.3. Calculation of blank corrected activity of the aliquot (A_c)

A_c (dpm) = A_a - V_a, where

V_a = the activity of the blank (dpm).

5.4. Calculation of uncertainty associated with the blank corrected activity of the aliquot (σ_{Ac})

$$\sigma_{Ac} = \sqrt{\sigma^2{}_{Aa} + \sigma^2{}_{Va}}\text{, where}$$

σ_{Va} = the uncertainty associated with the activity of the blank value.

5.5. Calculation of fractional recovery (R)

$$R = \frac{\left[\frac{c_{st}}{ts} - \frac{c_{bt}}{tb}\right]}{B_t \cdot A_{0t} \cdot E} \cdot 60\frac{s}{min} \cdot e\left[\frac{\ln 2}{T_{t,1/2}} \cdot (d_d - d_c)\frac{1 \text{ year}}{365.25 \text{ days}}\right]$$

5.6. Calculation of decision limit for aliquot (L_{dA})

$$L_{dA}(\text{dpm}) = \frac{3.29 \cdot \sqrt{\frac{c_b t_s}{t_b}\left(1 + \frac{t_s}{t_b}\right)} + 3}{B_s \cdot E \cdot R \cdot t_s}\left(60\frac{\text{dpm}}{\text{Bq}}\right)\text{, where}$$

E = The efficiency of the detector (Counter Eff.).

5.7. Calculation of sample concentration result (S_R)

$$S_R = \frac{A_c \cdot W_s}{W_a \cdot W_x}\frac{1000\text{ g}}{1\text{ kg}}\frac{1\text{ Bq}}{60\text{ dpm}}\text{, where}$$

A_c = Blank corrected activity of the aliquot (dpm)
W_s = Total solution weight at time of preparation (g)
W_a = Weight of aliquot used for analysis (g)
W_x = Wet weight analyzed if soft tissue, or ashed weight analyzed if bone (g), or equal to 1 (unit less) if radiochemical sample total activity to be calculated

The units are determined by the 'Tissue Type' and the 'Method' columns. For bone tissue analyzed via radioanalysis, the units are $\frac{\text{Bq}}{\text{kg of ashed weight}}$, and for 'soft' tissue, $\frac{\text{Bq}}{\text{kg of wet weight}}$, or $\frac{\text{Bq}}{\text{Radiochemical Sample}}$ for total isotopic activity in the radiochemical sample.

5.8. Calculation of uncertainty associated with the sample result σ_{SR}

$$\sigma_{SR}(\text{Bq / kg}) = \frac{W_s \cdot \sigma_{Ac}}{W_a \cdot W_x} \cdot \frac{1000\text{ g}}{1\text{ kg}} \frac{1\text{Bq}}{60\text{ dpm}}, \text{ where}$$

σ_{Ac} = Uncertainty associated with the blank corrected activity of the aliquot (dpm)

5.9. Calculation of concentration equivalent (C_E) for sample result

$$C_E\ (\text{Bq / kg or Bq / sample}) = \frac{W_s \cdot L_D}{W_a \cdot W_x} \cdot \frac{1\text{Bq}}{60\text{dpm}} \cdot 1000 \frac{\text{g}}{\text{kg}}$$

6. Sample calculations for absolute analysis

6.1. Calculation of sample activity (A_s)

$$A_S\,(\text{dpm}) = \frac{\left(\frac{C_s}{t_s} - \frac{C_b}{t_b}\right) \cdot 60(\text{S / min})}{B_s \cdot E}$$

6.2. Calculation of uncertainty in sample activity σ_{As}

$$\sigma_{As}\,(\text{dpm}) = \left[\frac{3600}{B_s{}^2 E^2}\left(\frac{c_s}{t_s{}^2} + \frac{c_b}{t_b{}^2}\right) + \frac{A_s^2}{E^2}\sigma_E{}^2\right]^{1/2}$$

6.3. Calculation of the limit of detection for sample activity L_{Ds}

$$L_{DA}\,(\text{dpm}) = \frac{3.29 \cdot \sqrt{\frac{c_b t_s}{t_b}\left(1 + \frac{t_s}{t_b}\right)} + 3}{B_s E t_s}\left(60\,\frac{\text{dpm}}{\text{Bq}}\right)$$

7. Literature Sources

7.1. ICRP 38. International Commission on Radiological Protection. Radionuclide Transformations: Energy and Intensity of Emissions.

7.2. Browne 1986. Brown, E.; Firestone, R.B., Shirley, V.S. (editor). Table of Radioactive Isotopes. John Wiley & Sones, New York. 1986.

7.3. ANSI N13.30. Performance Criteria for Radiobioassay. 1996.

7.4. Alphavision Software Reference Manual, Revision D. Software Version 4.02.

7.5. Currie, L.A. Limits for Qualitative Detection and Quantitative Determination. Application to Radiochemistry. Anal. Chem. 40(3), 1968. Pp 586-593.

7.6. LA-10300-M, Health and Environmental Chemistry: Analytical Techniques, Data Management, and Quality Assurance.

Table 2
Primary α Emissions for Selected Radionuclides

Isotope	α (keV)	α (%)	α2 (keV)	α2 (%)	α3 (keV)	α3 (%)	α4 (keV)	α4 (%)	α5 (keV)	α5 (%)	α6 (keV)	α6 (%)	α7 (keV)	α7 (%)	α8 (keV)	α8 (%)	α9 (keV)	α9 (%)
^{210}Po	5297	100																
^{226}Ra	4784	94.45	4602	5.55														
^{228}Th	5423	72.7	5340	26.7	5211	0.4	5177	0.18	5138	0.05								
^{229}Th	5052	1.6	5050	5.2	4978	3.17	4868	6.0	4901	10.2	4845	56.2	4838	4.8	4814	9.31	4798	1.25
^{230}Th	4688	76.3	4621	23.4	4480	0.12	4438	.030	4372	.001								
^{232}Th	4010	77	3952	23	3830	0.20												
^{232}U	5320	68.6	5264	31.2	5137	0.28												
^{233}U	4825	84.4	4804	0.05	4796	0.28	4783	13.2	4754	0.16	4729	1.6	4701	0.06	4664	0.04		
^{234}U	4776	72.5	4724	27.5	4605	0.24												
^{235}U	4597	5.0	4556	4.2	4414	2.1	4395	55	4370	6	4364	11	4344	2	4324	4.6	4216	5.7
^{238}U	4196	77	4147	23	4040	0.23												
^{237}Np	4873	2.6	4818	2.5	4804	1.6	4788	47	4772	25	4766	8	4707	1.0	4665	3.32	4640	6.18
^{236}Pu	5768	68.1	5721	31.7	5614	0.18												
^{238}Pu	5499	71.6	5456	28.3	5358	0.10												
^{239}Pu	5156	73.2	5143	15.1	5105	10.6												
^{240}Pu	5168	73.5	5124	26.4														
^{242}Pu	4901	78	4856	22.4														
^{241}Am	5544	0.34	5512	0.20	5486	85.2	5443	12.8	5388	1.4								
^{243}Am	5350	0.16	5319	0.12	5277	88	5234	11	5180	1.1								
^{244}Cm	5805	76.4	5763	23.6														

Reference: Browne 1986
Only principal emissions shown.

Table 3
Alpha Spectroscopy Region of Interest Analysis Parameters

				ROI Information (standard setting)			
Isotope	Half life* (years)	Primary α peak (keV)	Expected Primary α Peak Channel	FWHM (keV)	Left (channels)	Right (Channels)	Effective Branch Ratio
^{210}Po	378.9×10^{-1}	5297	359	50	22	8	1.0
^{226}Ra	1601	4784	257	50	45	8	1.0
^{228}Th	1.913	5423	385	50	50	8	1.0
^{229}Th	7340	4845	269	50	20	49	1.0
^{230}Th	7.54×10^{4}	4688	238	50	42	10	1.0
^{232}Th	1.405×10^{10}	4010	102	50	44	8	1.0
^{232}U	68.9	5320	364	50	40	8	1.0
^{233}U	1.592×10^{5}	4825	265	50	27	8	1.0
^{234}U	2.454×10^{5}	4776	255	50	38	8	1.0
^{235}U	7.037×10^{8}	4395	180	50	31	36	1.0
^{238}U	4.468×10^{9}	4196	140	50	36	8	1.0
^{236}Pu	2.851	5768	454	50	38	8	1.0
^{238}Pu	87.75	5499	400	50	32	8	1.0
^{239}Pu	2.411×10^{4}	5156	331	50	32	8	1.0
^{240}Pu	6564	5168	334	50	32	8	1.0
^{242}Pu	3.763×10^{5}	4901	280	50	32	8	1.0
^{241}Am	432.8	5486	397	50	23	18	1.0
^{243}Am	7380	5277	355	50	25	18	1.0
^{244}Cm	18.11	5805	461	50	45	10	1.0
^{237}Np	2.14×10^{6}	4788	258		90	25	1.0

*Half life information from Browne 1986.
Expected channel based on E (keV) = 3500 + 5 (keV/channel) * (Channel #)
^{229}Th provides a spectral interference for ^{230}Th at a rate of 2.7E-3/count ^{229}Th

Figure 1

Alpha Spectra of Typical Radionuclides

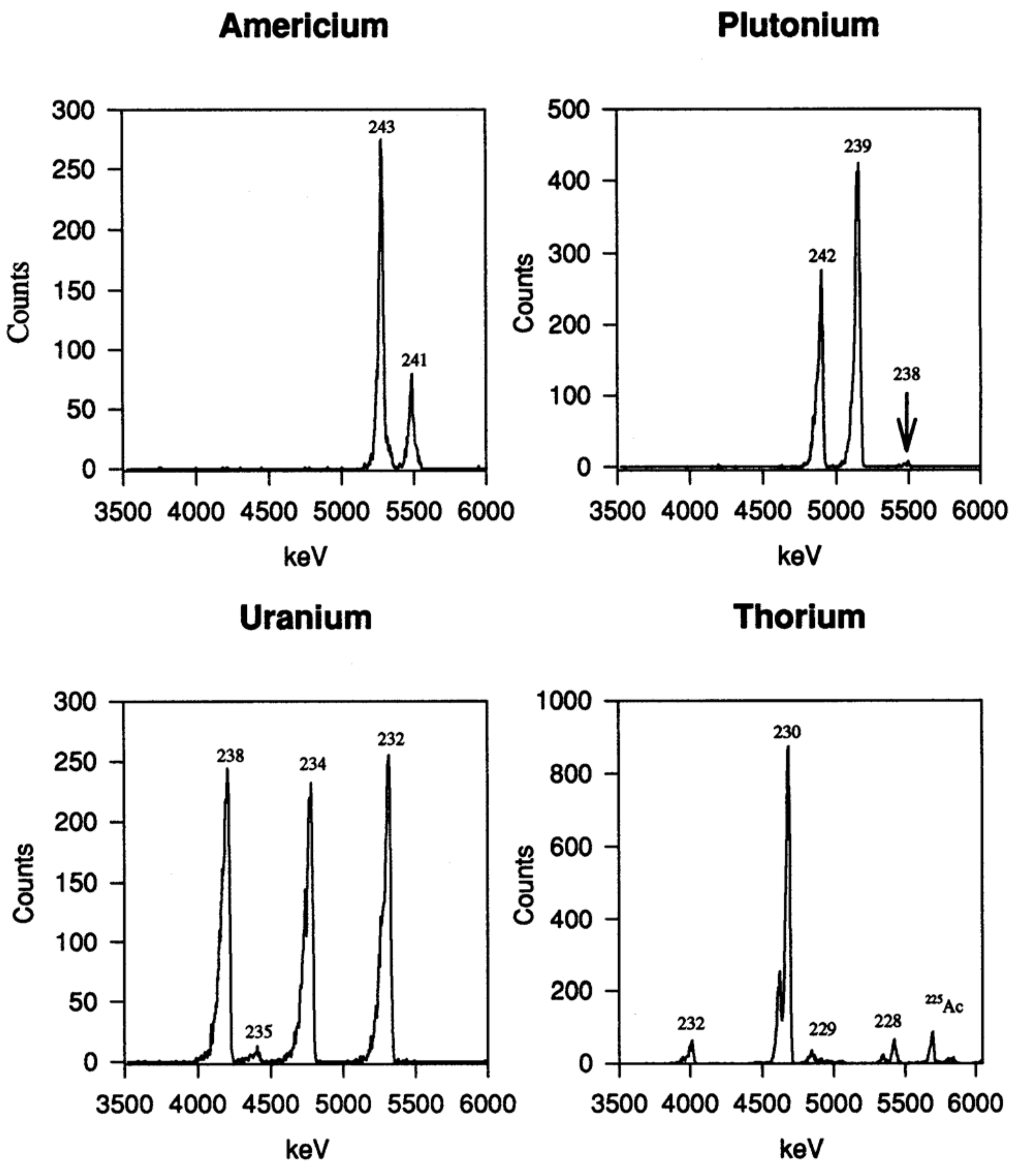

Figure 2
L_d as a Function of Time

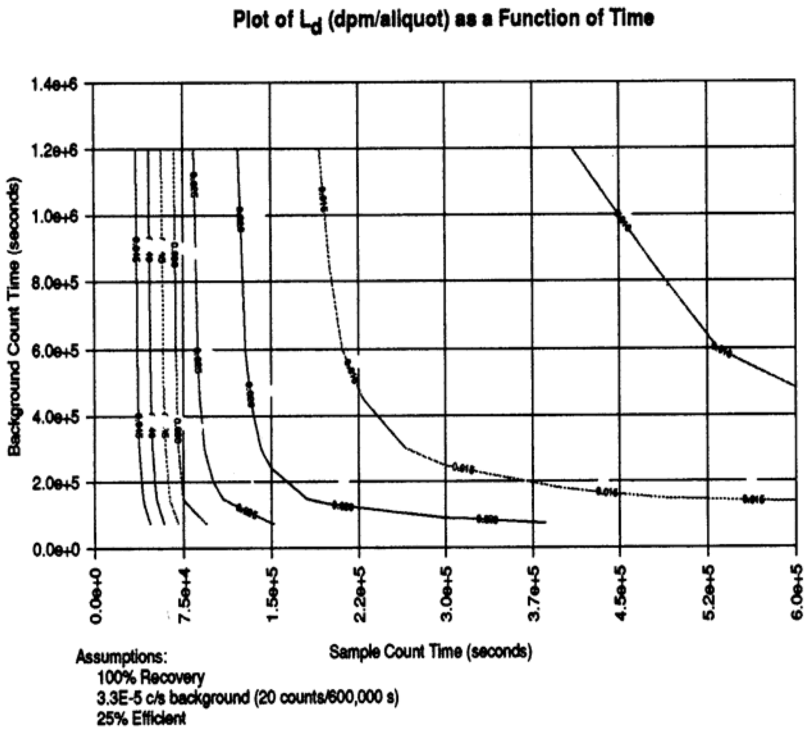

Figure 3
Relative Precision for 1 dpm Aliquot for Variable Recovery and Count Time

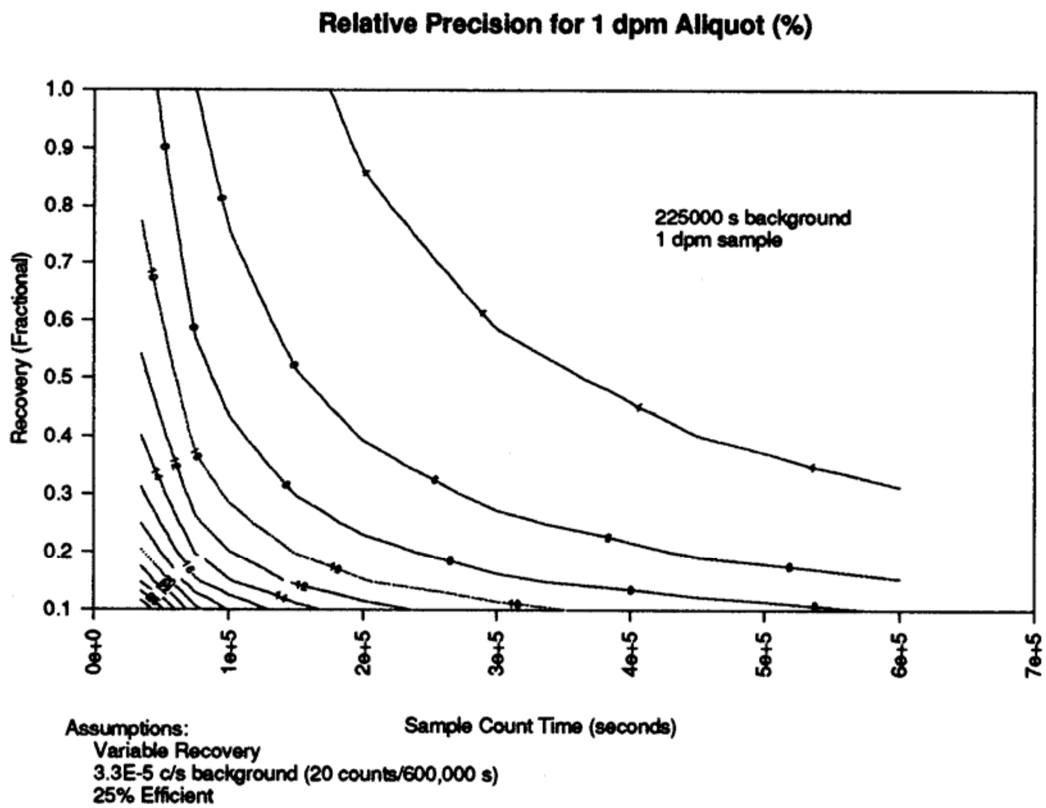

Figure 4
Relative Precision for 0.1 dpm Aliquot for Variable Recovery and Count Time

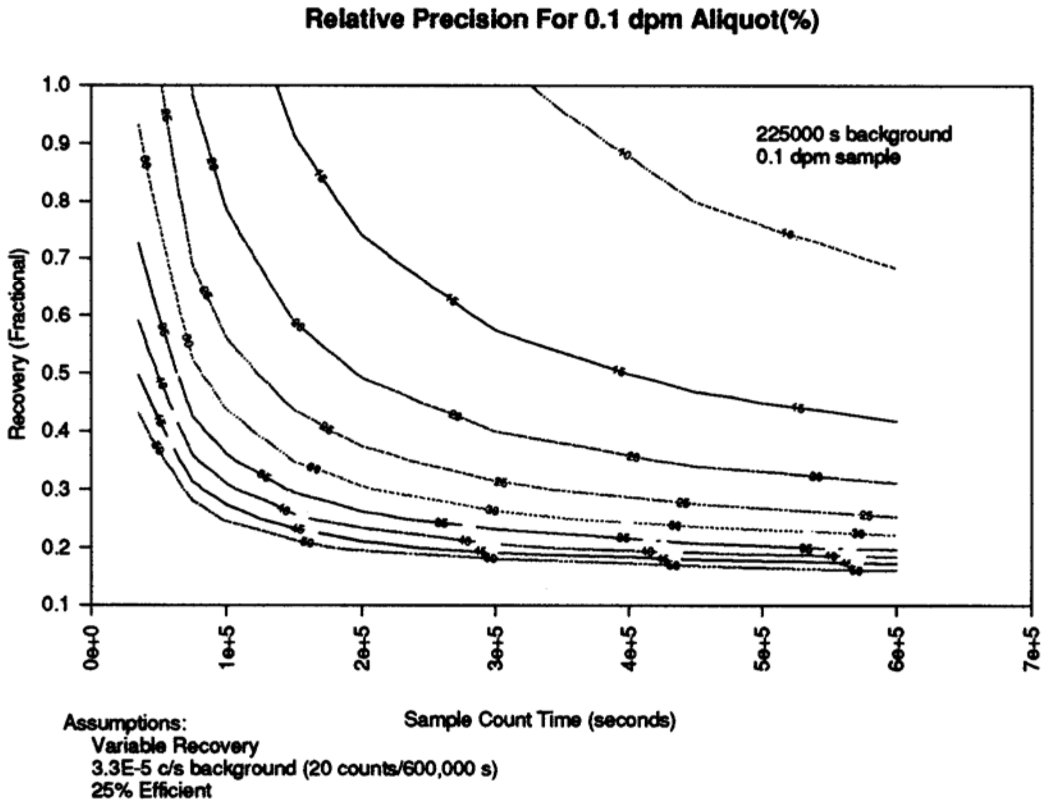

Figure 5
Relative Precision for 70% Recovery as a Function of Count Time and Aliquot Activity