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Optimization of Screening for Radioactivity in Urine by Liquid Scintillation

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Optimization of Screening for Radioactivity in Urine by Liquid Scintillation

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Abstract

Numerous events have or could have resulted in the inadvertent uptake of radionuclides by fairly large populations. Should a population receive an uptake, valuable information could be obtained by using liquid scintillation counting (LSC) techniques to quickly screen urine from a sample of the affected population. This study investigates such LSC parameters as discrimination, quench, volume, and count time to yield guidelines for analyzing urine in an emergency situation. Through analyzing variations of the volume and their relationships to the minimum detectable activity (MDA), the optimum ratio of sample size to scintillating chemical cocktail was found to be 1:3. Using this optimum volume size, the alpha MDA varied from 2100 pCi/L for a 30-second count time to 35 pCi/L for a 1000-minute count time. The typical count time used by the Sandia National Laboratories Radiation Protection Sample Diagnostics program is 30 minutes, which yields an alpha MDA of 200 pCi/L. Because MDA is inversely proportional to the square root of the count time, count time can be reduced in an emergency situation to achieve the desired MDA or response time. Note that approximately 25% of the response time is used to prepare the samples and complete the associated paperwork. It was also found that if the nuclide of interest is an unknown, pregenerated discriminator settings and efficiency calibrations can be used to produce an activity value within a factor of two, which is acceptable for a screening method.

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NOMENCLATURE

CDC	Centers for Disease Control and Prevention
cpm	counts per minute
dpm	disintegrations per minute
LEB	low-energy beta
LSC	liquid scintillation counting
MDA	minimum detectable activity
QC	quality control
RPSD	Radiation Protection Sample Diagnostics
SNL	Sandia National Laboratories
TRPDA	time-resolved pulse decay analysis
TSIE	transformed spectral index of an external standard
UGX™	Ultima Gold (Extended Range)™

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1. INTRODUCTION

1.1 Background

Numerous events have or could have resulted in the inadvertent uptake of radionuclides by fairly large populations. The technology associated with these events has (like all advances in technology) produced both positive and negative results. Some positive examples include:

- Deeper understanding of subatomic physics.
- The radio-pharmacology of tracers and cancer treatments.
- Tools for mineral prospecting and nondestructive industrial testing.
- Interplanetary exploration.
- Food and spice preservation.
- Nuclear energy.
- The expedited end of World War II and the nuclear deterrence of the Cold War.

A potential negative consequence of this technology is that concentrated radioactive materials have become ubiquitous in our time. It is conceivable that persons with ill intent may obtain radioactive material for use in terrorism. Such terrorism may result in uptakes of radioactive material by sizeable populations. Should a population receive an uptake, valuable information could be obtained by quickly screening the urine of a sample of the population. Some example questions to be answered by this screening include:

- Are radionuclides present or not present in the urine?
- If radionuclides are present, what are the activity levels (concentrations)?

The Centers for Disease Control and Prevention (CDC) is considering these questions as a part of its counterterrorism and homeland security activities. The Radiation Protection Sample Diagnostics (RPSD) program at Sandia National Laboratories (SNL) has been analyzing urine for radiological dosimetry purposes utilizing the technology of liquid scintillation counting (LSC) since 1995. The CDC funded this study performed by SNL to leverage the RPSD work of the past to determine the most appropriate and effective ways for the CDC to apply LSC in the immediate aftermath of a terrorist-caused or accidental release of unknown radioactive material that affects a large population. This study was approved by SNL's Human Studies Board.

1.2 The Liquid Scintillation Counting Process

An overview understanding of the LSC process is necessary to understand the study designed and performed by SNL's RPSD program to address the CDC requirements. In the RPSD program, a sample is usually mixed with a chemical cocktail that scintillates (produces light) in the presence of alpha or beta radioactivity. The characteristics of the scintillation can reveal much about the radioactivity. Most easily, analysis of the scintillation can determine whether the radioactivity is due to an alpha or beta emitter. Less easily, the scintillation can roughly measure the energy of an emitted particle, a possible signature of the nuclide(s) present.

Discrimination is a key to identification and quantification of the nuclide(s) present or not present. The LSC machines commercially available often have an adjustable discriminator (alpha/beta separation) as an option. RPSD uses the Tricarb line of LSC machines. Another key consideration is the cocktail and its behavior in contact with the sample. This is known in LSC radiological measurement as quench. In other words, the chemistry of the sample affects both the ability of the cocktail to scintillate and how it scintillates. Thus quench depends on the chemical makeup of the sample. In the case of urine, the chemical makeup of samples can vary widely from person to person and for a particular person from day to day. Quench is critical to the identification and quantification of the nuclide(s) present or not present. In general, quench effects make quantification and identification difficult. Also in general, larger volume samples enhance the ability to detect radioactivity, but the enhancement at some point will be offset by quench effects. Finally, longer count duration enhances the ability to detect radioactivity, but at the expense of the rate of sample through-put.

This overview provides a minimal understanding of the process. For more detail, please refer to Appendices A and B.

1.3 The Liquid Scintillation Counting Study

Most users of LSC machines know beforehand what nuclides they expect to be present (or not present) in samples. Therefore users tend to calibrate their instruments to respond optimally to nuclides of their concern. At the request of the CDC, this study concentrated on alpha emitters Plutonium-239, Curium-244, and Thorium-230 in addition to beta emitters of various energies. The study does not involve radiochemical separations; therefore mixtures of multiple beta and/or alpha emitters were not assessed in detail.

In response to the CDC requirement to obtain information quickly about the radiological characteristics of the urine from a sample of a population, the RPSD program at SNL designed and performed a study of LSC that varied the following parameters:

- Discrimination.
- Quench.
- Volume.
- Count duration.

Studying these parameters yielded guidelines for analyzing urine for radionuclides by liquid scintillation. The guidelines are intended to help the CDC users balance the speed of obtaining useful information against other considerations, especially the minimum detectable activity (MDA). To develop these guidelines the following relationships were analyzed:

- Optimum discriminator setting as a function of multiple combinations of alpha and beta energies.

- Quench values encountered for typical urine samples through a study of historic samples.

- Quench as a function of sample volume for typical urine.

- MDA as a function of sample volume.

- MDA as a function of sample count time.
- Time requirements for both sample preparation and analysis.
- Effect on reported sample activity found when varying discriminator settings away from optimal.

In the course of this study, SNL's RPSD program performed the following tasks for the CDC:

1. Determined fifteen optimum discriminator settings for multiple alpha/beta combinations: alpha emitters Pu-239, Cm-244 and Th-230 each versus beta emitters H-3, Ni-63, Cs-137, Cl-36 and Sr/Y-90 each. These results are presented in Part 3 and Appendix C.
2. Determined quench value distribution for urine by studying historical data gathered by RPSD to identify key information that defines 'typical' urine. These results are presented in Part 2.
3. Determined counting efficiency of the LSC versus quench for two alpha- and beta-emitting radionuclide combinations, Pu-239/Sr-90 and Cm-244/Cs-137. These results are presented in Part 4.
4. Using typical urine as defined by Task 2 above, determined a relationship between sample quench and sample volume. These results are presented in Part 4.
5. Using water, determined relationship between sample volume and MDA. These results are presented in Part 4.
6. Using the relationships from Tasks 4 and 5 above, attempted to determine a relationship between sample volume of typical urine and MDA. These results are presented in Part 4.
7. Using typical urine and a practical volume based on Task 6 above, determine a relationship between MDA and count time. These results are presented in Part 5.
8. Analyzed capabilities for quantifying nuclides when discriminator settings are other than optimum. These results are presented in Part 4.
9. Performed analyses to estimate probable time requirements for sample receipt, analysis, and reporting. These results are presented in Part 5.

In addition to the study tasks listed above, RPSD personnel will provide CDC staff with on-site training in the theories of alpha- and beta-emitting nuclide discrimination and the calibration techniques RPSD utilizes. These theories and techniques may or may not be adaptable to the instrumentation the CDC is currently using. No software or special modifications to the RPSD spreadsheets for use with the CDC's current LSC machines were developed through this study.

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2. LIQUID SCINTILLATION QUENCH VALUE DISTRIBUTION TO DEFINE TYPICAL URINE

As part of its internal radiation dosimetry program, SNL has collected hundreds of urine samples per year since 1995. The vast majority of these samples are analyzed to verify the absence of significant amounts of radioactive material.

One way to perform these analyses is LSC. In the SNL program one part urine is mixed with three parts of LSC cocktail. SNL uses Perkin-Elmer's Ultima Gold (Extended Range)^{TM} or UGXR^{TM}, a nontoxic biodegradable cocktail. UGXR^{TM} is an acceptable cocktail for this purpose in that it allows a large aqueous sample loading and behaves well with other natural chemicals found in urine. The samples are counted in Perkin-Elmer's liquid scintillation counter model 2500 with the alpha/beta discrimination option.

Because LSC is the measurement of light coming from the cocktail with the light related to the amount and character of the radioactivity present, interference with that light will affect efficiency. This interference is known as *quench*. Quench can be caused by chemical interference (especially pH) or color/opacity.

Efficiency in this context is the ability of a system to detect radiation from a sample. It is measured as the ratio of detected radioactive disintegrations to the number of radioactive disintegrations that have occurred in the sample over some time period. Efficiency is discussed in greater detail in Part 4.

The chemical makeup of urine can vary widely from person to person and from the same person day to day. Because of this variability, chemical and color quench also will vary widely from sample to sample. SNL therefore measures the quench present in each individual sample during the counting process. This measurement, coupled with a quench calibration, allows calculation of the approximate value of a sample's efficiency.

Quench is measured by placing a standard gamma source next to each sample. The gamma photons cause scintillation in the sample by producing Compton electrons. The amount of light collected is measured against a standard scale known as the *transformed spectral index of an external standard* or TSIE. In SNL's Perkin-Elmer instruments, TSIE is an arbitrary scale of 0-1000 where 0 is total lack of detected scintillation and 1000 is the scintillation of a 20-ml vial of totally unquenched and optimally fluor-doped toluene.

As a prerequisite to other experiments, SNL wanted to determine typical quench in urine samples from a large population. Rather than collect many new samples, SNL reviewed results from its internal dosimetry program. This program mixes 5 ml of urine with 15 ml of UGXR^{TM}. Figure 1 shows the measured quench distribution for approximately 2400 historical samples, with TSIE values rounded to the nearest ten.

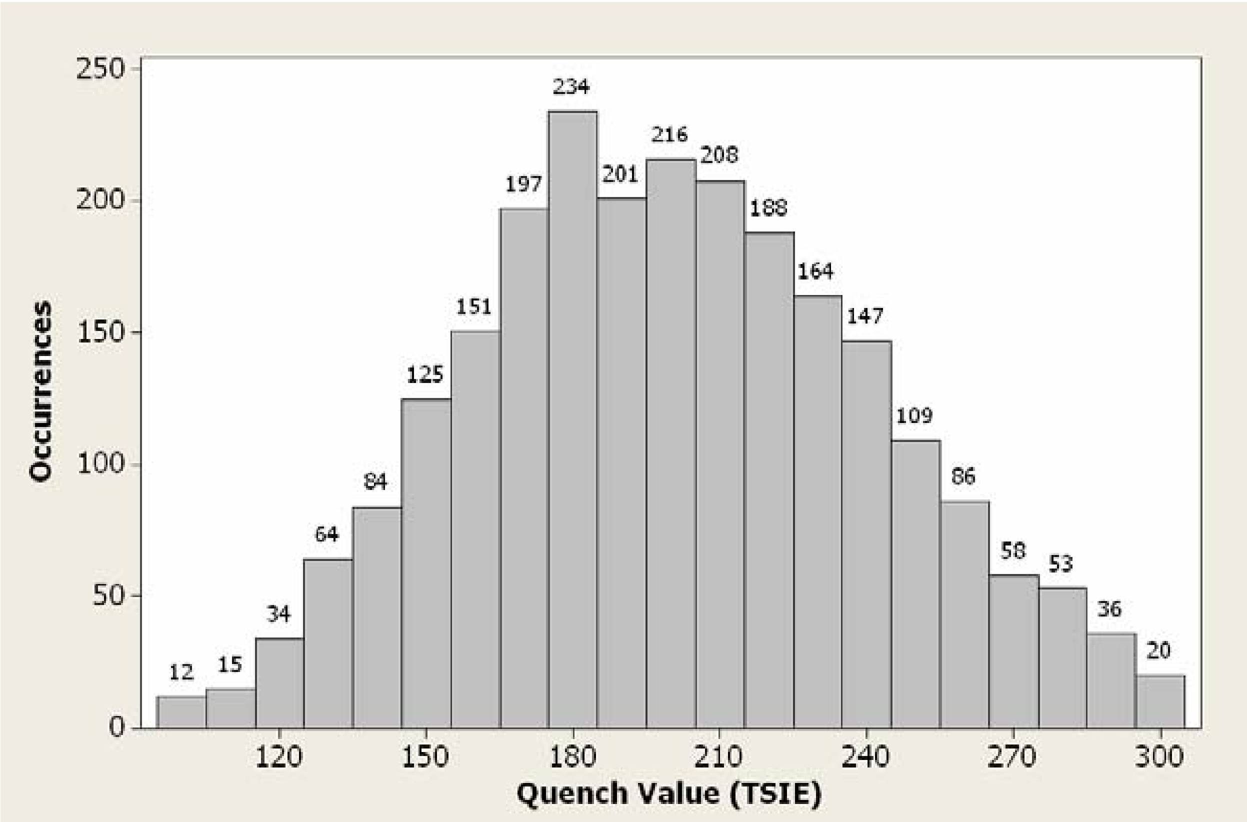

Figure 1. Historical sample quench measures

Figure 1 shows a roughly normal distribution for quench. Figure 2 uses an overlaid Gaussian curve to show that a 5-ml urine aliquot in 15 ml UGXR^{TM} scintillation sample results in a TSIE of 200 ± 40 about two-thirds of the time (one standard deviation). For further experiments urine that results in a quench value within this range is defined as *typical*.

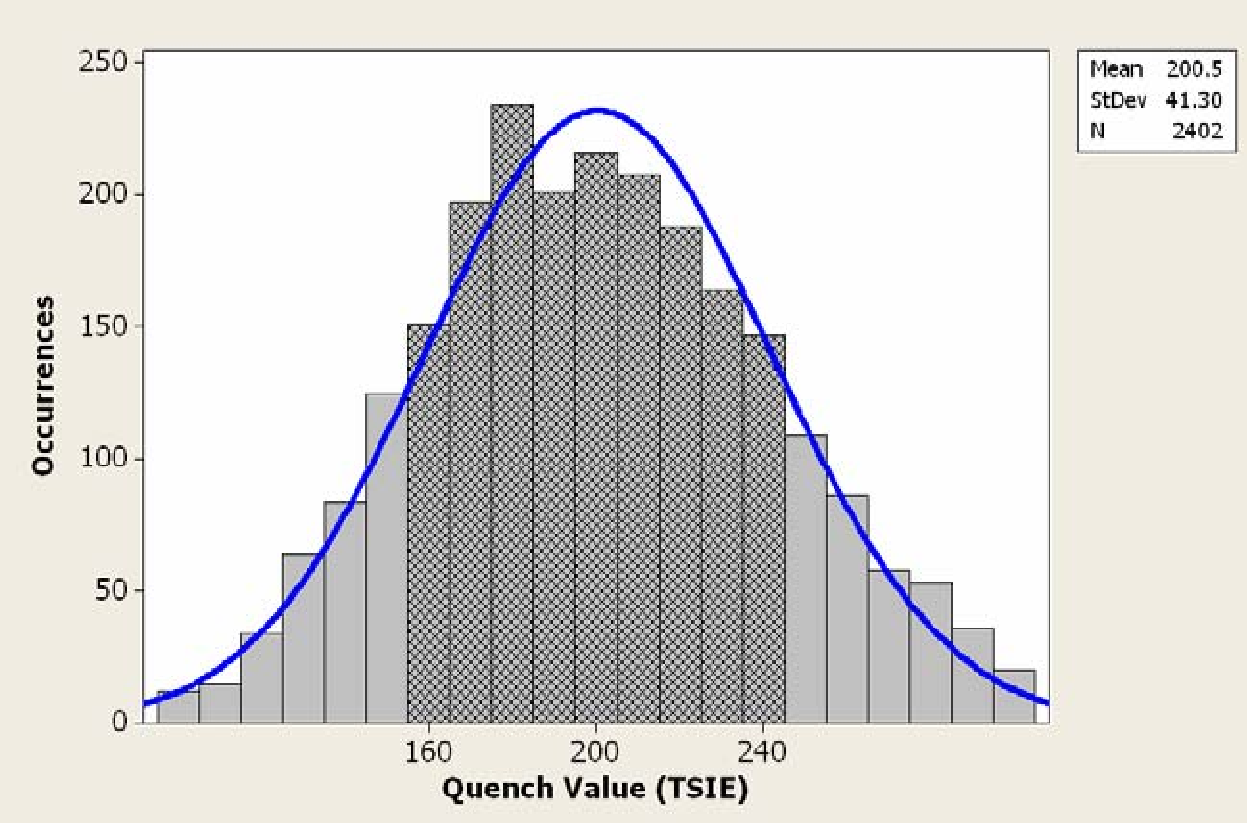

Figure 2. Urine in UGXR™

The typical quench derived from the 2400 historical samples could be affected by the following four considerations:

1. These samples were not all analyzed on the same instrument. TSIE can vary slightly for a particular sample from instrument to instrument. TSIE also can vary slightly for different batches of vials and scintillation cocktail.

2. These samples were collected for SNL's internal dosimetry project from 2002 to 2006. Most are periodic samples from the same individuals several times over that period.

3. The participants in SNL's internal dosimetry project almost all reside in the same geographic area: the Albuquerque, New Mexico metropolitan area. It is likely that the chemical makeup of the samples is slightly affected by this geographic homogeneity.

4. Almost always, the persons who supplied these samples had ample advance knowledge that the samples were requested. Unlike an emergency screening situation, the participants supplied the samples at their convenience. It is possible that the chemical makeup of the samples is affected by whether the provision of the sample was well-anticipated or if it was provided on short (emergency) notice.

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3. OPTIMUM DISCRIMINATOR SETTINGS FOR VARIOUS COMBINATIONS OF ALPHA- AND BETA-EMITTING RADIONUCLIDES

SNL used a Perkin-Elmer brand liquid scintillation counter (Tricarb model 2500TR, serial number 419272) to study 15 combinations of alpha- and beta-emitting nuclides to determine optimum discriminator settings for each combination. The 15 combinations included three alpha-emitting nuclides of various monoenergies and five beta-emitting nuclides of various maximum energies. The LSC machine used has the alpha/beta discrimination option, which uses time-resolved pulse decay analysis (TRPDA) to determine whether a detected radiation event is due to an alpha-emitting nuclide or a beta-emitting nuclide. This discrimination tool allows simultaneous screening for both alpha- and beta-emitting nuclides.

The discriminator is not (and cannot be) perfect, in that sometimes an alpha-emitting nuclide is misidentified as a beta-emitting nuclide and vice versa. Such misidentifications are variously known as *crosstalk*, *spillover*, or *overlap*.

3.1 The Radionuclides

Table 1 lists the nuclides studied along with their associated particle energies.

Table 1. Radionuclide and Associated Particle Energies

Alpha Emitters		Beta Emitters	
Nuclide	**Energy (keV)**	**Nuclide**	**Maximum Energy (keV)**
Th-230	4700	H-3	19
Pu-239	5150	Ni-63	67
Cm-244	5800	Cs-137	514
		Cl-36	710
		Sr/Y-90	2190

Alpha emitters tend to be heavy nuclei with decay energies between 4000 and 6000 keV. Beta emitters span a wide range of energy and, due to concurrent neutrino emission, are not mono-energetic.

The Tricarb 2500TR instrument has an arbitrary discrimination scale of 256 possible settings. The 15 combinations of alpha- and beta-emitting nuclides resulted in a wide range of settings. But it was found that low-energy beta (LEB) emitters (Ni-63 and H-3) would rarely be misidentified as alpha emitters at any practical setting. These practical settings are those at which higher energy betas have a significant chance of being misidentified as alphas. Higher energy beta emitters would be misidentified as alpha emitters at most 10% of the time across this middle range of discriminator settings. Conversely, alpha emitters would be misinterpreted as betas 10% of the time across that same range.

Pu-239 and Cl-36 are a typical combination, in that their decay energies are mid-ranged. This combination resulted in a discriminator setting of 108. Figure 3 shows the discrimination calibration graph generated to determine the setting value, where total alpha plus beta spillover is minimized. Appendix C provides the discriminator calibration graphs for all 15 combinations considered in this study.

	% Spillover		
Setting	Alpha	Beta	Total
0	3.3%	84.6%	88.0%
18	3.8%	83.3%	87.1%
36	3.8%	82.3%	86.1%
54	3.9%	75.7%	79.6%
72	4.5%	65.2%	69.7%
90	5.3%	37.9%	43.1%
108	9.6%	9.2%	18.8%
126	20.5%	1.3%	21.8%
144	41.6%	0.5%	42.1%
162	66.3%	0.2%	66.5%
180	84.5%	0.1%	84.6%
198	93.4%	0.1%	93.5%
216	97.9%	0.1%	97.9%
234	99.3%	0.0%	99.4%
252	99.6%	0.0%	99.7%
Optimum:			
108	9.59%	9.21%	18.8%

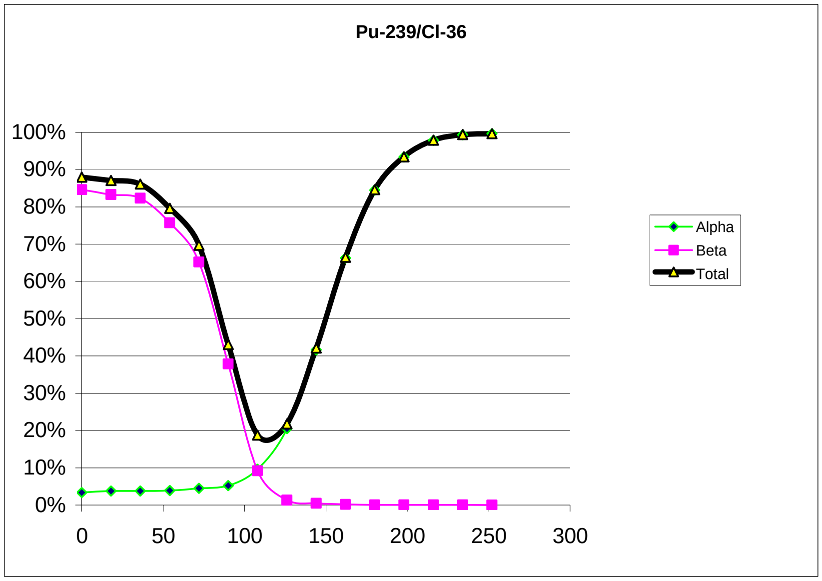

Figure 3. Discriminator setting (108) for Pu-239/Cl-36 combination

The 15 discriminator settings determined in this study are summarized in Figure 4. The higher energy beta emitters coupled with a range of alpha emitters resulted in discriminator settings in the narrow range (104 to 115):

		Alpha Emitters			
		Th-230	Pu-239	Cm-244	Maximum Beta Energy (keV)
Beta Emitters	H-3	90	92	93	19
	Ni-63	45	41	69	67
	Cs-137	104	105	108	514
	Cl-36	106	108	110	710
	Sr/Y-90	111	112	115	2280
		4700	5150	5800	
		Alpha Energy (keV)			

Figure 4. Discriminator settings

The least energetic beta with significant likelihood of being misinterpreted (Cs-137) coupled with the least energetic alpha (Th-230) resulted in a discriminator setting of 104. Conversely, the most energetic beta (Sr/Y-90) coupled with the most energetic alpha (Cm-244) resulted in a discriminator setting of 115 on the scale of 0 to 256. (Again, the settings found when using the LEBs of Ni-63 and tritium (H-3) are neglected due to those nuclides having a very low likelihood of being misinterpreted as alpha emitters.) Thus even if the discriminator is set incorrectly for a given nuclide or mix of nuclides, SNL asserts that good screening would still be possible. This hypothesis is tested next.

3.2 Discriminator Settings Other Than Optimum

To study the effects of incorrect discriminator settings, known amounts of various nuclides or *spikes* were counted at several settings in the reasonable discriminator range (104-115). These spikes are commonly measured in the convenient activity units of disintegrations per minute (dpm).

First, with the discriminator setting at 110, which is the correct setting for a combination of Cm-244 and Cl-36 and the setting normally used by RPSD, the variety of spikes were counted to populate Table 2. Again, there are three alpha emitters with mono-energies 4700, 5150, and 5800 keV and beta emitters of maximum energy 19 to 2280 keV (see Figure 4).

Table 2. Activities (dpm) Measured with Discriminator at 110

Nuclide	Spike	Alpha	Beta	LEB	Total
Pu-239	1140	1160	90	40	1290
Cm-244	**1130**	**1120**	40	0	1160
Th-230	1070	1050	110	0	1160
H-3	980	0	0	1000	1000
Ni-63	1270	0	130	2410	2540
Cl-36	**1370**	100	**1390**	200	1690
Cs-137	1030	60	1120	480	1660
Sr-90	2230	160	1310	320	1790

The following observations can be made about Table 2:

- The nuclide measurement results in bold are closest to their respective spike values. This is expected because they are the nuclides used to determine the discriminator setting.
- The Pu-239 and Th-230 results (alpha emitters) are also very close to their spike values.
- The H-3 result (reported as LEB activity) is close to the spike value because SNL uses an assumption that betas detected below a certain energy (12 keV) are due to tritium.
- The Ni-63 result (mostly reported as LEB activity) has the greatest deviation from its spike value. This is because most of the Ni-63 betas are registered below 12 keV and counted as LEBs. Therefore a lower counting efficiency is assumed (discussed in Part 4)

and a higher calculated activity is the result. Nevertheless, even with the lower mistaken efficiency applied, the result is within a factor of two of the known spike value. In practice, if it is known that Ni-63 is the nuclide of interest, a strict Ni-63 efficiency calibration would be used. For gross screening, however, a factor of two absolute error is usually considered acceptable.

- The Cs-137 beta result is in good agreement with the spike. This is expected because Cs-137 and Cl-36 have maximum beta energies of comparable magnitude.

- Similarly to Ni-63, the Sr-90 result is incorrect in beta activity but within a factor of two. Sr-90 is particularly difficult to measure precisely, because it is usually found in secular equilibrium with its progeny Y-90. It is therefore a mixture of beta emitters with differing maximum energies. (To be specific, Sr-90 decays with maximum beta energy of 546 keV and its daughter Y-90 decays with maximum beta energy of 2280 keV).

- Finally, note that beta emissions misinterpreted as alpha activity are rare and vice versa, thus showing the reliability of TRPDA discrimination.

Tables 3 and 4 show results for the discriminator settings at 104 and 115 respectively. The nuclide combinations used to determine the settings and the spike measurement results are shown in bold.

Table 3. Activities (dpm) Measured with Discriminator at 104

Nuclide	Spike	Alpha	Beta	LEB	Total
Pu-239	1140	1200	30	50	1280
Cm-244	1130	1120	10	0	1140
Th-230	**1070**	**1090**	50	0	1140
H-3	980	0	0	1100	1100
Ni-63	1270	0	10	2560	2570
Cl-36	1370	170	1310	210	1690
Cs-137	**1030**	100	**1060**	510	1670
Sr-90	2230	30	1120	330	1480

Table 4. Activities (dpm) Measured with Discriminator at 115

Nuclide	Spike	Alpha	Beta	LEB	Total
Pu239	1140	1180	80	40	1300
Cm-244	**1130**	**1120**	40	0	1160
Th-230	1070	1030	120	0	1150
H-3	980	0	0	1120	1120
Ni-63	1270	0	120	2600	2720
Cl-36	1370	50	1450	210	1710
Cs-137	1030	40	1140	500	1680
Sr-90	**2230**	80	**1420**	320	1820

The Table 2 observations also apply to Tables 3 and 4. In each case, the discriminator setting is optimum for a particular alpha/beta combination. Therefore, the setting is not optimum for all other nuclides. However, an examination of these tables shows that a middle-range setting is adequate to perform screening in an emergency or other situation when the nuclide or mix of nuclides is not known.

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4. RELATIONSHIPS BETWEEN QUENCH, VOLUME, AND MINIMUM DETECTABLE ACTIVITY FOR TYPICAL URINE IN LIQUID SCINTILLATION

The concept of MDA is treated with much rigor in many other publications. For this study, MDA is used as a key measure of the optimization of sample and detector configurations.

Four parameters of concern affecting MDA are:

1. Sample size.
2. Detector size.
3. Quench.
4. Background.

Because these four parameters are interrelated in a complex way, an attempt was made to determine how they behave by studying them empirically. Carefully measured volumes of pure water and/or typical urine were mixed with likewise varying volumes of scintillating cocktail (with the sum volume kept constant). The results provide guidance to help optimize a screening process.

4.1 Minimum Detectable Activity in Context

In general and particularly in a screening process, the goal is to be able to detect small amounts of radioactive material in a sample in a short time and with a minimum effort. Reasonable, economical measures to reduce MDA and thus improve the results are generally desired.

MDA depends on several factors, among them the *efficiency* of the counting system. Efficiency in this context is the ability of a system to detect radiation coming from a sample. Efficiency is measured as the ratio of detected radioactive disintegrations to the actual number of radioactive disintegrations that have occurred in the sample. If measured over some time interval, it is the ratio of detected radioactivity to the radioactivity present. All other factors being equal, a larger efficiency results in a smaller MDA; i.e., MDA is inversely proportional to efficiency:

$$MDA \propto \frac{1}{\varepsilon}$$

where = efficiency.

Efficiency and therefore MDA also depend on the size of the detector used in the detecting system. A larger detector will more likely detect radiation from a sample, so a larger detector will result in a smaller MDA. In the extremes, for example, a detector of no size will detect no radiation and a detector of infinite size will detect all radiation. The relationship is correct only in the extremes, however. The efficiencies of intervening sizes can at best be modeled or calibrated.

MDA, when the 'A' actually stands for *specific* activity, depends on sample size. All other factors being equal, a larger sample size results in a smaller minimum detectable specific activity. For urine the sample size is usually measured as a volume (V):

$$MDA \propto \frac{1}{V}$$

In SNL's LSC program, the sample is mixed with the detector—the scintillating cocktail. For constant sample plus cocktail volume (total 20 ml), an optimum must exist, because while increasing sample size, detector size is simultaneously decreasing. Similarly, increasing detector size requires decreasing sample size.

As mentioned above, LSC is the measurement of light coming from the cocktail with the light related to the amount and character of the radioactivity present. Interference with that light is called *quench*. Quench is caused by chemical interference, coloring, opacity, and pH. Varying the size of a sample such as urine, which is laden with constituents that add quench, will affect efficiency and thus MDA.

Finally, there are always system imperfections and radioactivity not of interest. This 'noise' obscures the 'signal' and is known as *background*. Increasing background increases MDA, according to the following relationship.

$$MDA \propto \sqrt{N_b}$$

where N_b = number of background counts detected.

4.2 Sample Size/Detector Size

In this test the total volume of the scintillator/sample was held constant at 20 ml. Deionized water was varied from 0.0 to 20.0 ml in 0.5-ml increments with the balance being UGXR™ scintillating cocktail. This range obviously includes extremes in which there is no sample or insufficient cocktail to dissolve or interact with the sample.

TSIE was used as the single indicator of quench for these combinations. When using UGXR™ cocktail the TSIEs ranged from 360 for pure UGXR™ cocktail to near zero (essentially no response), as shown in Figure 5. For this study, all other factors were assumed to be equal and contiguous batches of scintillation vials and cocktail were used.

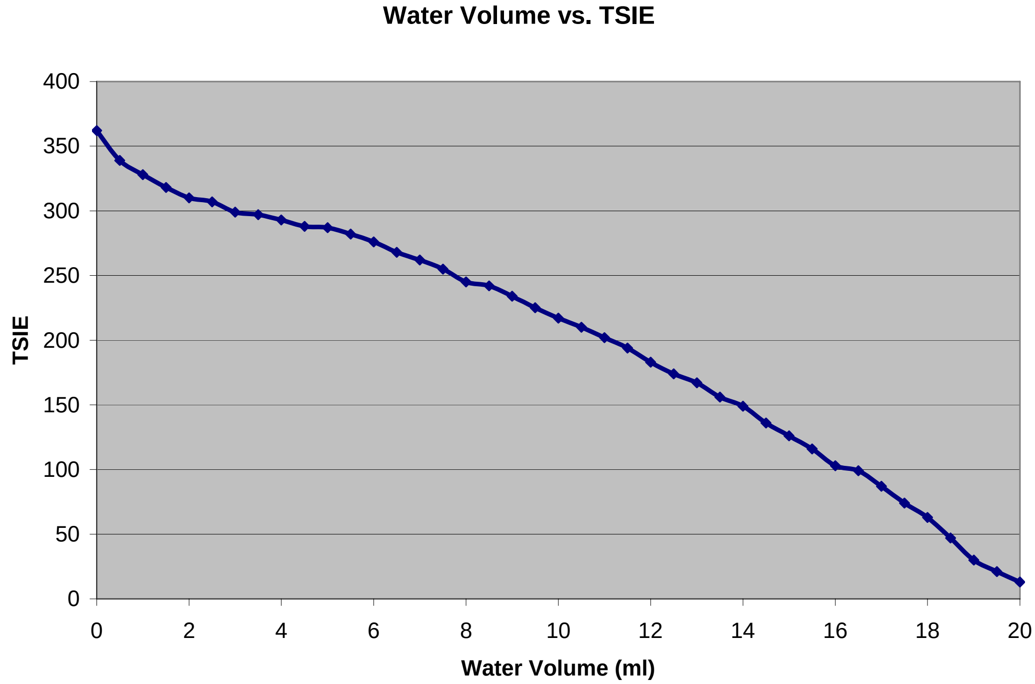

Figure 5. Relationship between sample volume and TSIE

TSIE falls below 100 at a water volume of 16 ml. This is also the point at which the water and cocktail failed to dissolve and phased layers were observed. Thus with phasing the only consideration, the maximum sample volume is 16 ml.

Also, a fairly linear relationship was observed across the range of volumes, but with a noticeable flattening between 2 and 8 ml, as shown in Figure 6.

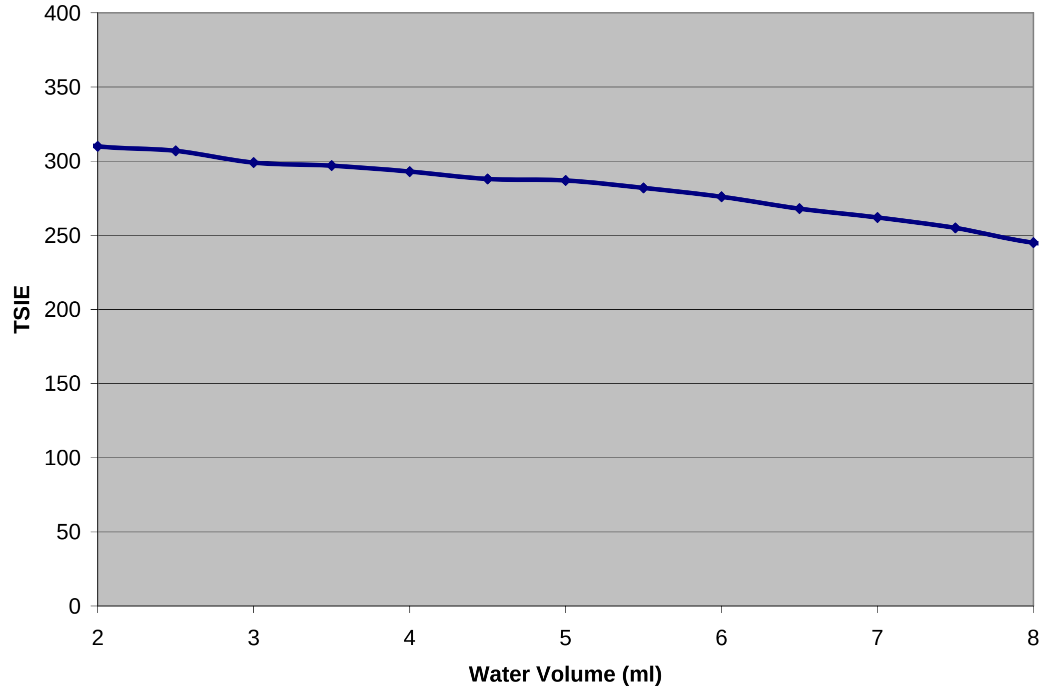

Figure 6. Water volume vs. quench

This region has a high TSIE (low quench) and will result in acceptable efficiency values for detecting higher energy betas and typical alphas, as shown below. This level relationship is desirable because changes in sample volume in this range do not result in proportional changes in TSIE.

4.3 TSIE versus MDA

Figure 7 shows that below a TSIE of 200, efficiency drops steeply for higher energy betas and typical alphas.

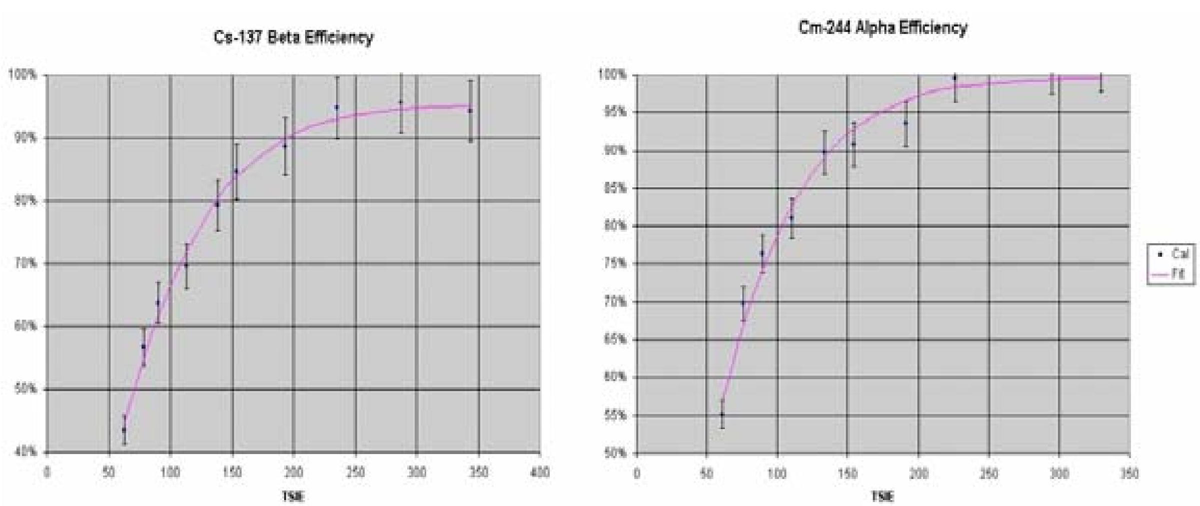

Figure 7. Efficiency for higher energy betas and typical alphas

Efficiency becomes unacceptably low below 100, especially when screening for LEB emitters such as tritium, as shown in Figure 8.

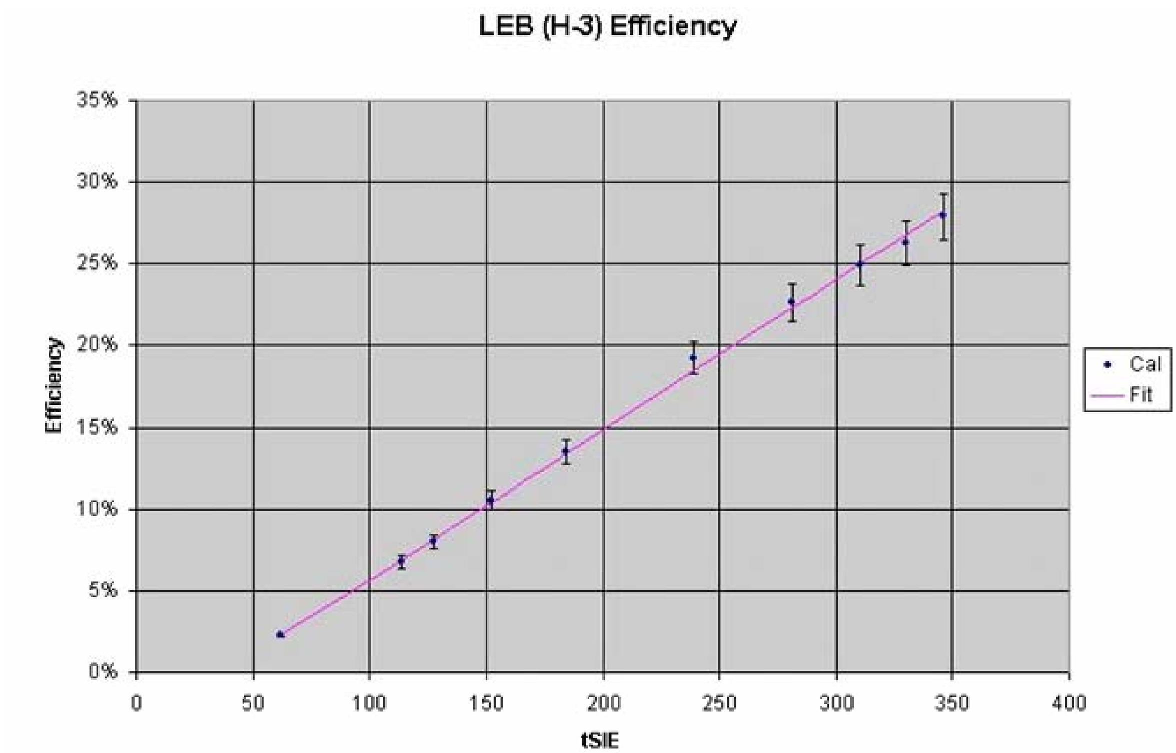

Figure 8. LEB (H-3) efficiency

A general guideline for these observations and information is that sample loading based on TSIE alone allows up to 15 ml water and 5 ml cocktail. This results in a TSIE of 125. Applying this to the calibration curves in Figures 7 and 8, the efficiencies obtained are about 85% for alphas, 75% for higher-energy betas, and 8% for low-energy betas such as tritium. Experience has shown, however, that any introduction of chemicals other than water reduces the sample's solubility and increases quench. Obviously, urine screening includes chemicals other than water.

As discussed previously, when increasing the sample size, the detector size is simultaneously decreasing. This is because the sample and detector are in intimate contact at a constant volume of 20 ml. Thus in order to determine a relationship between sample volume and MDA, simplifying assumptions must be made. Beginning with the following relationship, from L. Currie[1] and adopted in NUREG/CR-4007[2]:

$$L_c = k\sigma_0$$

where

$$\sigma_0 = \frac{\sqrt{2N_b}}{t}.$$

L_c = critical limit (or critical level or decision level/limit), in counts observed in a time *t* for detecting a randomly occurring phenomenon such as radioactivity. The critical limit in this context is the highest value of activity reportable by a counter for a sample with no activity (to a high degree of confidence—usually set to 95%).

k = a multiplier that for normal (Gaussian) distributions of random events specifies the degree of confidence. For 95% confidence, *k*=1.645.

$_0$ = the standard deviation (error) of the normal distribution of a sample with no activity.

N_b = the number of background counts found in the acquisition time *t*.

Figure 9 is a graphical representation of this concept.[3]

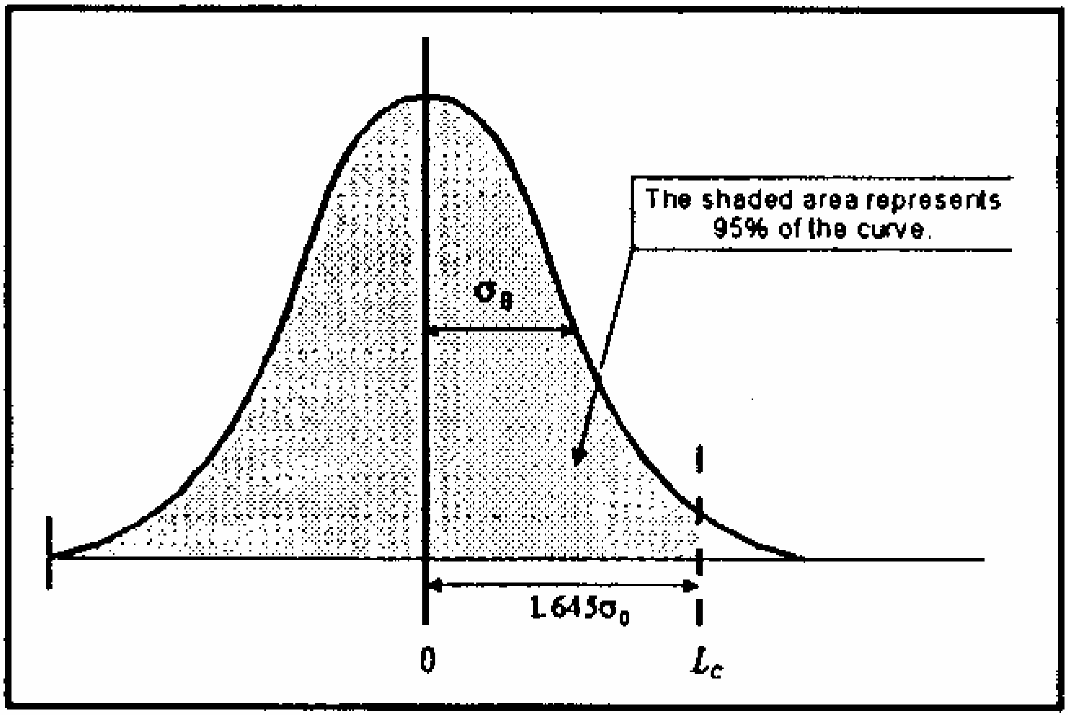

Figure 9. Critical limit of detection with 95% confidence

In this example, 95% of samples with zero activity will be reported below the critical limit (including negative values).

To convert L_c into an activity measurement, divide by efficiency ( ), which in LSC is a function of quench and the character of the radiation of interest. To convert to a *specific activity*, divide by volume *V* (or other measurement types such as mass or area):

$$L_c = \frac{k\sigma_0}{\varepsilon V}$$

Consider the efficiency (quench) curve for Cs-137, shown in Figure 10.

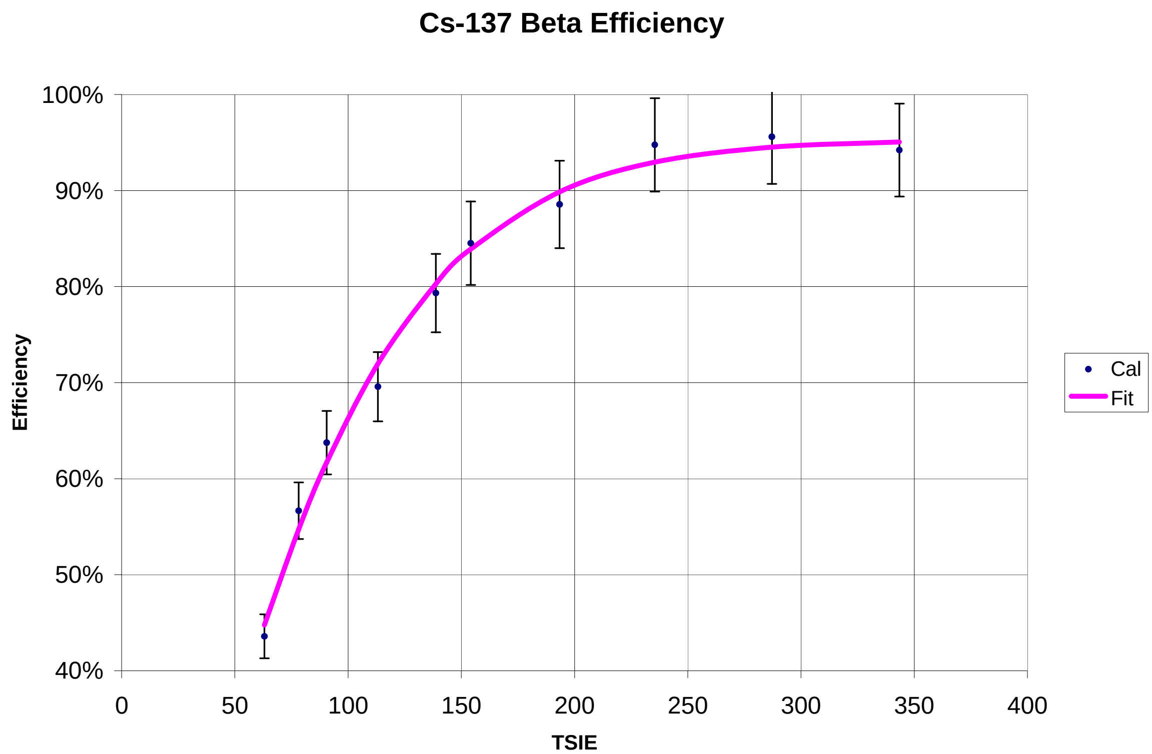

Figure 10. Efficiency curve for Cs-137

The data points are produced by adding spikes of single value and a quenching agent (nitromethane) in varying amounts to the scintillation cocktail. The curve is generated by least-squares fit conveniently by the natural empirical equation:

$$\varepsilon = \varepsilon_0 - e^{-P1*TSIE^{P2}}$$

where

ε_0 = the efficiency found in an unquenched spike.

*P*1 and *P*2 are fitting parameters.

The resulting best fit efficiency equation for the curve used in Figure 10 is:

$$\varepsilon = 0.95 - e^{-0.0032*TSIE^{1.3}}$$

As an example, consider that the TSIE for a particular sample of water mixed with UGXR™ was measured using the external gamma source to be 100. Visually using Figure 10 or by calculating using the equation above, the efficiency for measuring Cs-137 activity in this sample is about 68%. In other words, of every 100 disintegrations of Cs-137 occurring in the sample, 68 should be detected.

Combining these concepts yields:

$$L_c = k\frac{\sqrt{2N_b}}{t\varepsilon V}$$

Assume that a typical background count rate is 40 counts per minute (cpm) and is measured over a 30-minute period to yield a total of 1200 counts. For this example (converted to commonly used specific activity units for water):

$$L_c = (1.645)\left(\frac{\sqrt{(2)(1200)}\ \text{counts}}{30\ \text{minutes}}\right)\left(\frac{100\ \text{disintegrations}}{68\ \text{counts}}\right)\left(\frac{1}{0.005\ \text{liter}}\right)\left(\frac{1\ \text{minute}}{60\ \text{sec}}\right)\left(\frac{10^{12}\ \text{pCi - sec}}{3.7\times10^{10}\ \text{disintegrations}}\right)$$

L_c = 360 pCi/L for Cs-137 to a (traditional) 95% confidence.

To consider MDA, restate its common definition: MDA is the activity that, if present in a sample, provides a certain confidence (again, usually 95%) that the activity will be reported above the L_c. Figure 11 is a graphic representation of this concept.[3]

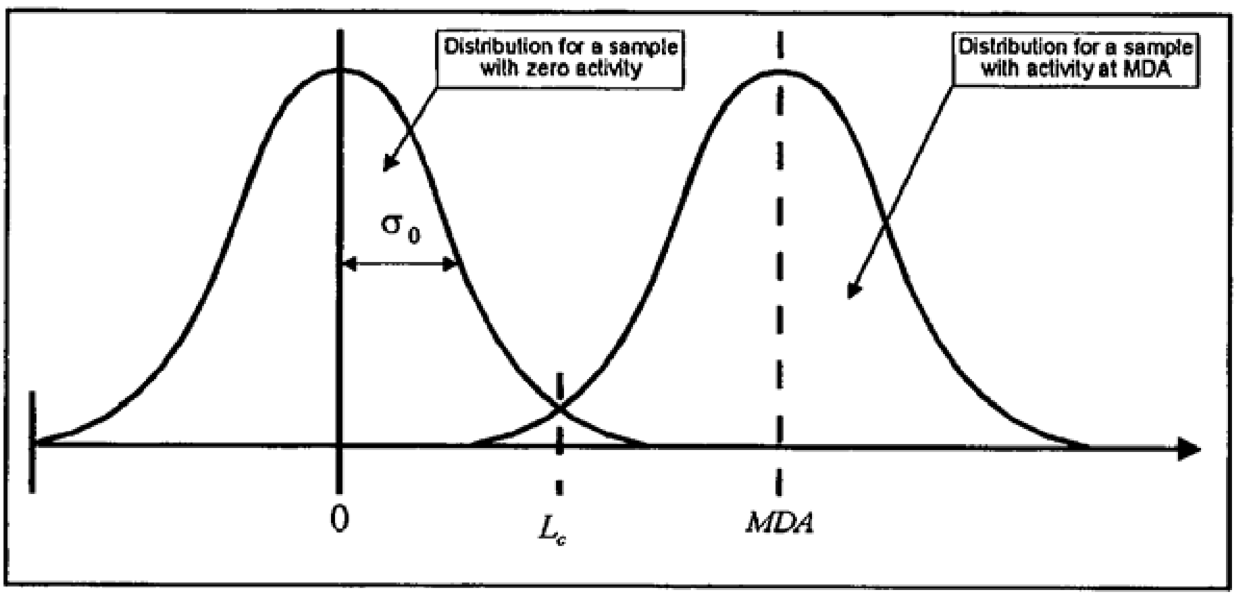

Figure 11. Error distributions for samples with no activity and for samples with MDA

Mathematically this is expressed as:

$$MDA = \frac{k^2}{t\varepsilon V} + 2L_c$$

It can be shown that with nominal count times *t*, efficiencies , and volumes *V*, that the first term, $\frac{k^2}{t\varepsilon V}$, reduces and can be disregarded in practice. Thus for simplicity, as shown graphically in Figure 11:

$$MDA \approx 2L_c$$

In the water example above, then, MDA   720 pCi/L at the traditional confidence.

With these explanations, it is now possible to examine the relationship of MDA to volume of sample while simultaneously varying detector size (volume of scintillation cocktail). Using the calibration recorded above and from data obtained empirically by SNL, the following parameters emerge:

1. LEB (such as tritium) efficiency:

$$\varepsilon_{LEB} = 0.98 - e^{-.00032 * TSIE^{1.2}}$$

With LEB background of 507 counts in 30 minutes, the relationship of MDA to increasing sample volume (simultaneously to decreasing cocktail volume) is shown in Figure 12.

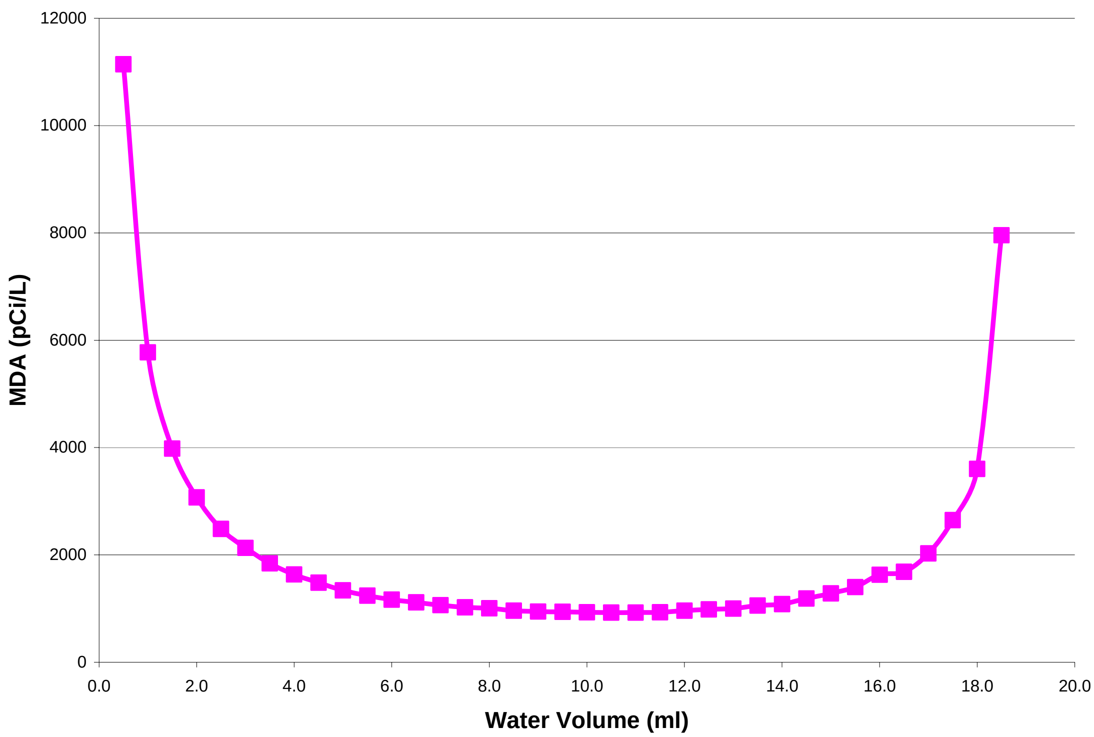

Figure 12. MDA for low-energy betas

2. Higher energy betas (such as Cs-137) efficiency:

$$\varepsilon = 0.95 - e^{-0.0032 * TSIE^{1.3}}$$

With higher energy betas and a background of 1190 counts in 30 minutes, the relationship of MDA to increasing sample volume is shown in Figure 13.

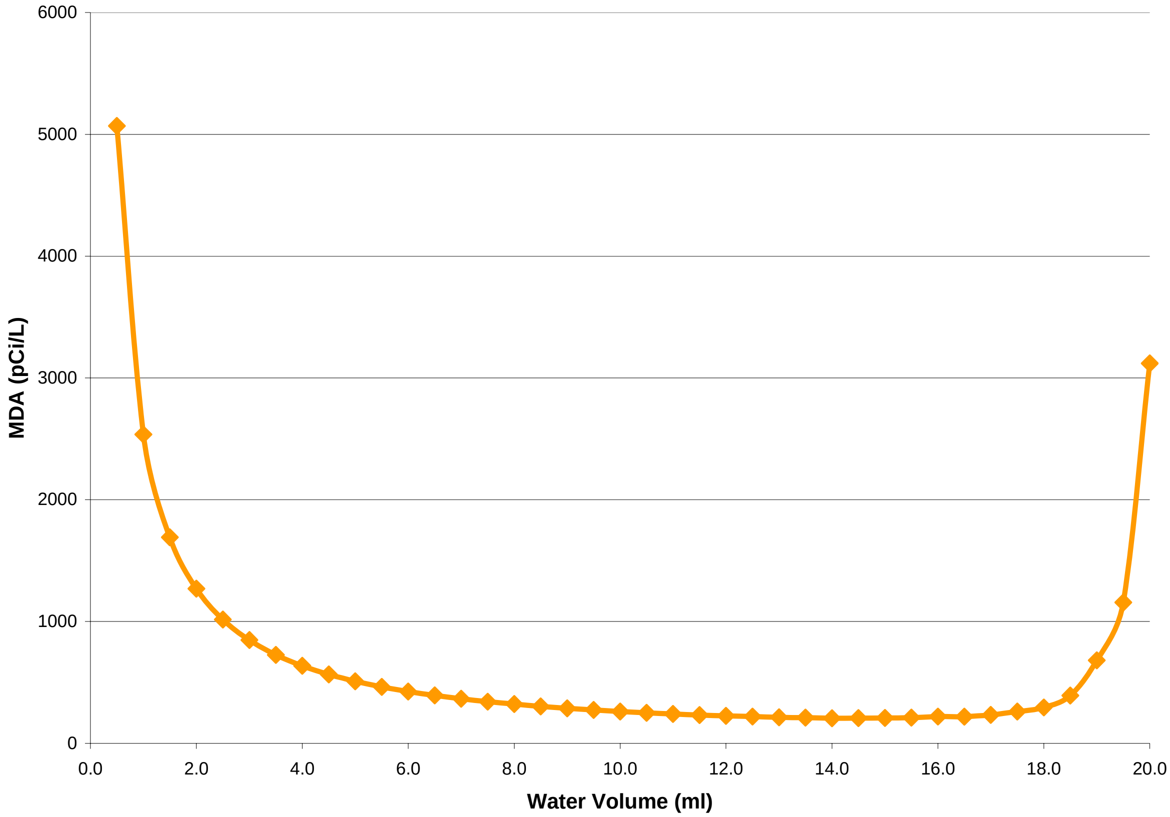

Figure 13. MDA for higher energy betas

3. Alpha emitters (such as Cm-244) efficiency:

$$\varepsilon = 1.0 - e^{-0.0053 * TSIE^{1.2}}$$

With alpha emitters and a background of 228 counts in 30 minutes, the relationship of MDA to increasing sample volume is shown in Figure 14.

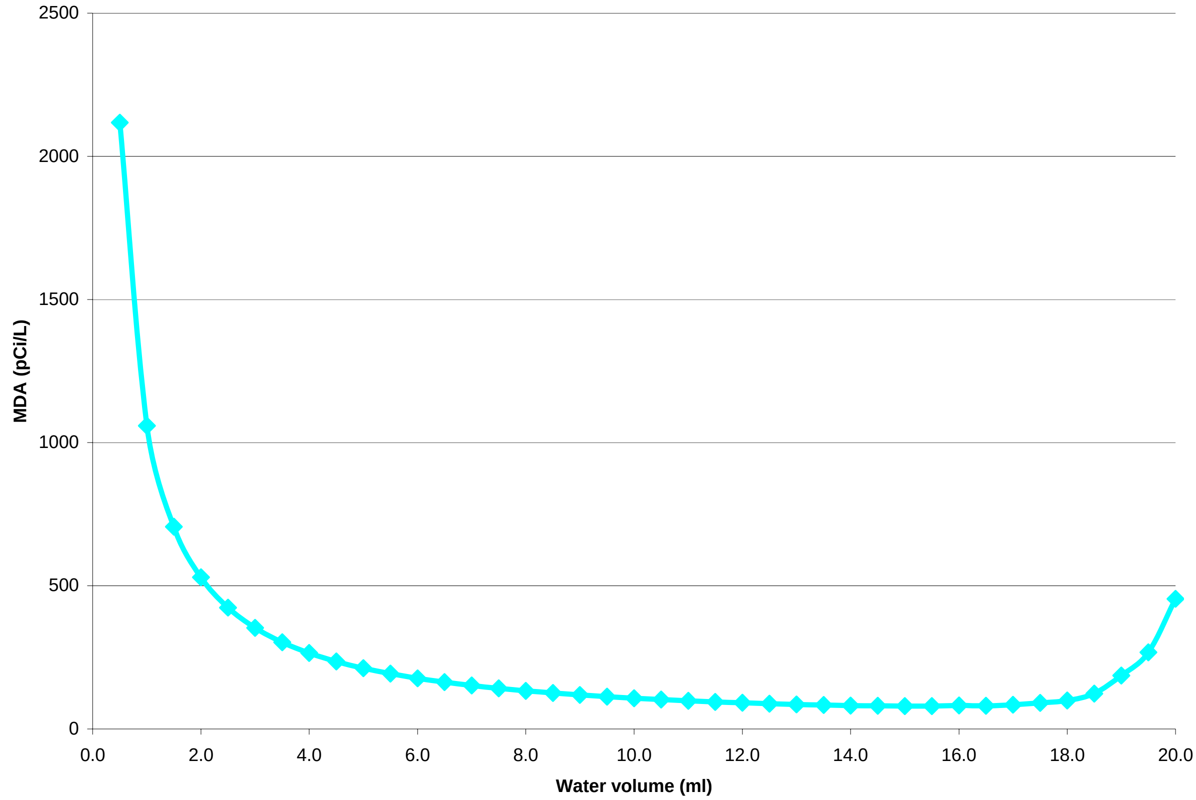

Figure 14. MDA for alphas

In each case, the same general shape is observed and would be observed with marginal differences in background counts: a steep decline in MDA as sample volume increases from 0.5 ml to 3 ml. This is followed by a certain flat response with a diminishing improvement on the increasing sample volume (decreasing detector size) until 16 ml. After 16 ml the system quickly fails with a steep increase in MDA. The flatness can be termed a *buffering effect*, defined as the possibility of significant deviations in sample size away from the optimum sample size while MDA is relatively constant. Figure 15 combines the relatively flat portions of the curves from Figures 12 through 14.

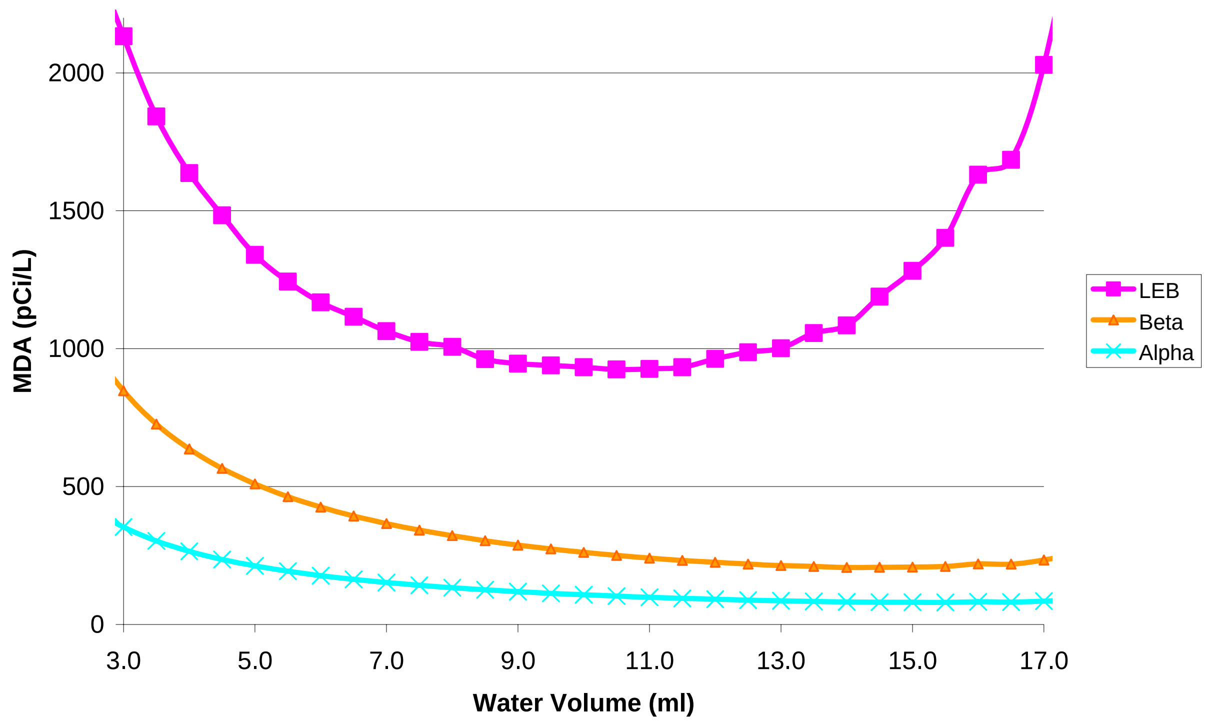

Figure 15. Sample volume vs. MDA

A minimum (optimized) MDA of 900 pCi/L for LEB was observed with a sample size equal to detector size at 10 ml. Also noted is that MDAs for higher energy betas and alphas are constant across this wide range of volumes. From 5 ml to 10 ml, LEB MDA improves nearly 30% from 1300 pCi/L to its minimum. Along this same variation, higher energy beta MDA improves roughly 50%, but absolute MDA changes by 250 pCi/L. Likewise alpha MDA changes only about 100 pCi/L, which is a 50% improvement.

4.4 Application to Urine Screening

The implication of the observations described is that for screening relatively clean samples such as water, the optimum mixture of sample and detector (cocktail) is 1:1, but that the absolute return sacrificed when using smaller sample loading is not difficult to accept because the detector simultaneously becomes a larger, more efficient size.

The exercise was repeated using typical urine, with the knowledge that chemicals other than water are present in the samples.

In this test, the total volume of the scintillator/urine sample was again kept constant at 20 ml. The volume of typical urine was varied in 0.5-ml increments with the balance being UGXR™ scintillating cocktail until the mixture phased or failed to combine, as shown in Figure 16.

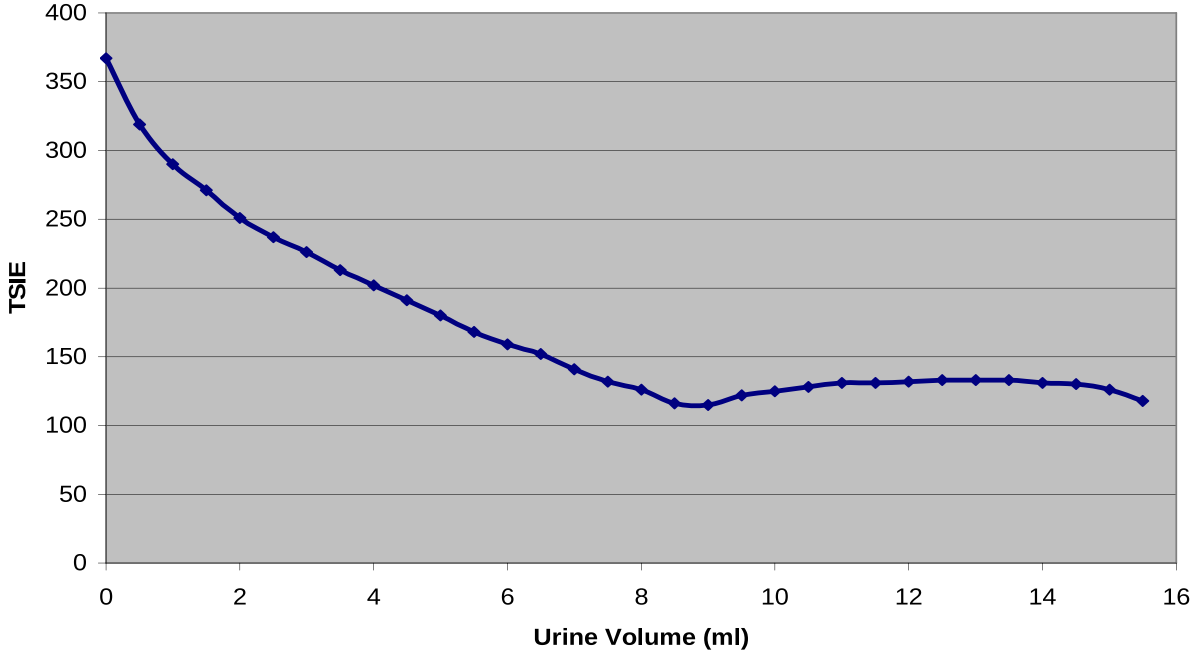

Figure 16. Urine volume vs. quench

At 9 ml typical urine (11 ml UGXR™), the mixture failed to combine. When that happens, geometrical relationships within the sample/detector system are considered invalid: i.e., the efficiency calibration does not apply.

Applying these TSIE values for the same background conditions, count times, and efficiency curves as previously applied for water, the relationships illustrated in Figure 17 were revealed.

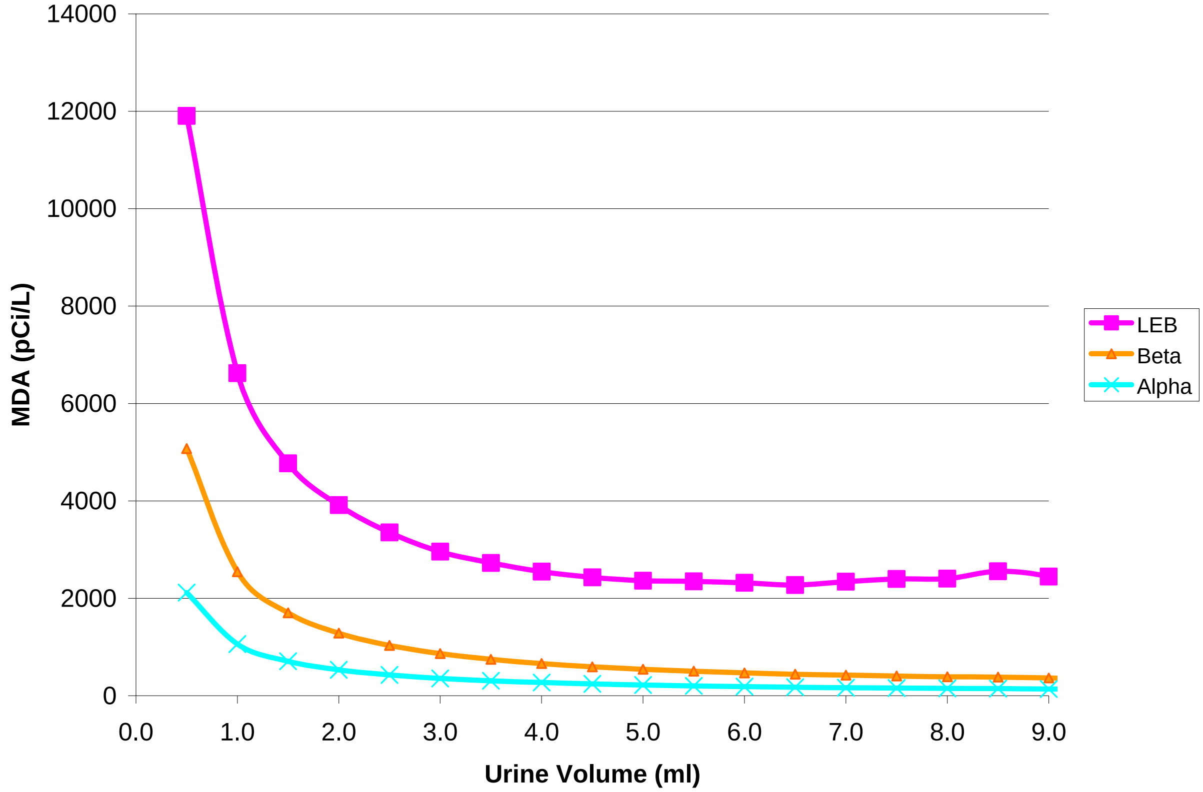

Figure 17. Sample volumes vs. MDA

MDAs for volumes between 3 and 7 ml urine (balance UGXR™) are fairly flat functions. Small gains are available in relative terms by using the very maximum of this range, but minimal in absolute terms of activity as shown in Figure 18.

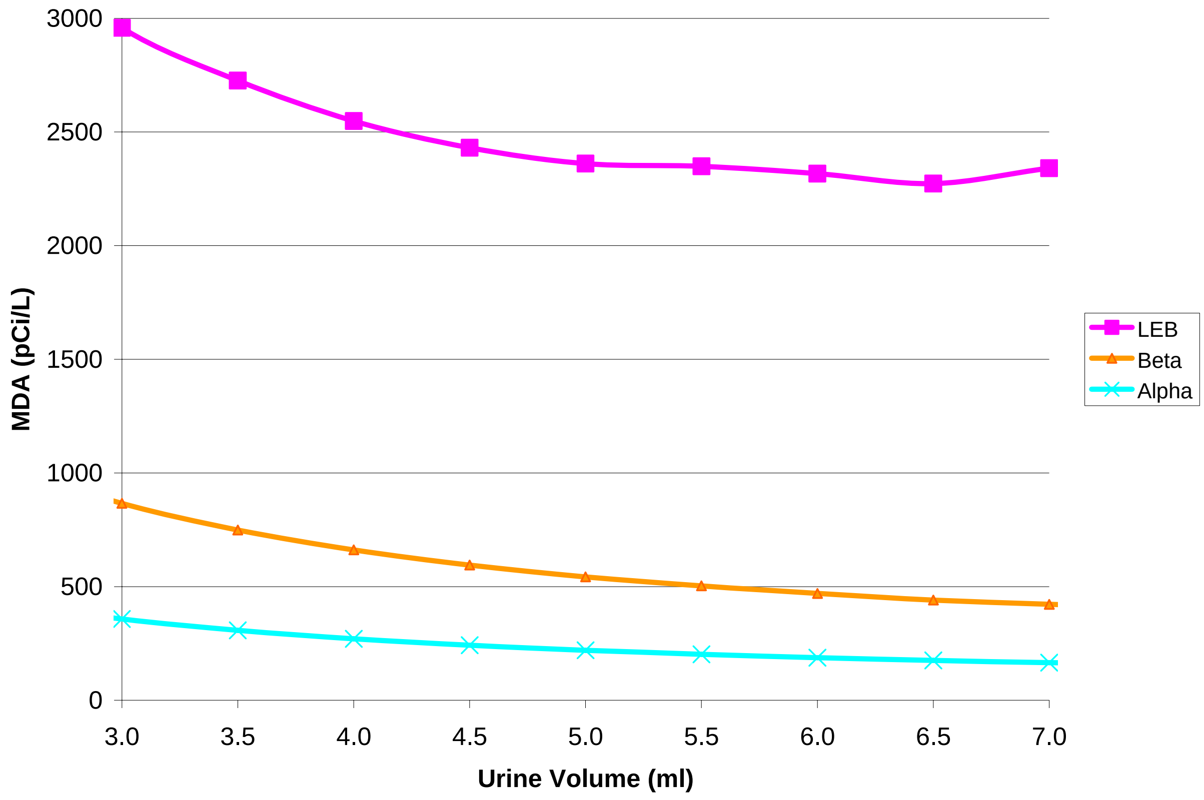

Figure 18. Sample volume vs. MDA

4.5 Optimum MDA Results

This study suggests selecting a midpoint for sample size in the urine volume vs. MDA function: i.e., 5 ml urine plus 15 ml UGXR™. This is one part urine with three parts scintillation cocktail. While never before studied in this way to determine the optimum ratio, RPSD settled on this ratio based on long experience. SNL and CDC now know that this is a desirable ratio in that:

1. It is in the midpoint between a useful MDA at the low end and phase separation (calibration invalidation) at the high end.

2. Volume variations intrinsically affect MDA inversely, but also affect efficiency inversely and thus MDA proportionally in LSC. This is seen in the buffering property (flatness of the function on each side of the chosen aliquot).

3. For typical urine it will result in acceptable quench and thus efficiency. Urine is deemed typical for this purpose as resulting in TSIE of 200 ± 40 in one standard deviation (two-thirds) of samples. Other (atypical) urine used in this sample/cocktail configuration results in TSIE of 200 ± 80 in 98% of samples—usually still acceptable for screening. Only 2% of samples should fall outside this range, with half of those approaching the quench of deionized water and the other half (only 1%) unacceptably quenched (TSIE too low).

4. As noted previously, urine contains potassium-40, adding to background. At a 5:15 configuration, it was found that this is not a significant hindrance to effective screening.

5. ADDITIONAL URINE SCREENING CONSIDERATIONS

This section of the study provides guidance on time considerations. These considerations include sample preparation time in urgent situations, relationship of count time to MDA, real-time analysis, and automatic statistical tests.

5.1 Emergency Screening Drill

Almost all urine samples screened for radioactivity through SNL's RPSD program are processed for routine purposes. They are collected periodically from individuals who work with and have some chance (usually remote) of ingesting radioactive material.

On one occasion, when RPSD received 30 samples for routine analysis, the batch was treated as if it were an emergency. The samples were processed in accordance with established procedures without sacrificing any quality control (QC), but with particular urgency and focus. The simulated emergency began at the time of sample receipt: i.e., all sample collection and cataloguing occurred before arrival at the RPSD laboratory. The steps performed and the time required for each step are discussed below.

1. ***Sample receipt (0.5 hour)***: Samples were logged into the RPSD accounting system. This step includes populating a sample request form and preliminary screening for radioactivity to determine handling precautions. (There were no particularly radioactive urine samples found based on the preliminary screening, so handling was deemed to be routine and relatively easy).

2. ***Miscellaneous preparatory work (1.0 hour)***: This involved unpacking the samples, pre-labeling sample vials, and checking instruments to be used, such as the LSC machine, balances, and pipettes.

3. ***Sample preparation (2.0 hours)***: The samples were aliquoted and mixed with cocktail. This step also includes preparing QC tracking samples, such as spikes and blanks.

4. ***Sample counting (16 hours):*** Each sample was counted for 30 minutes. This time can be drastically reduced by splitting samples between several counters if they are available.

5. ***Data processing (0.5 hours):*** Raw counting data was converted to reportable activity results.

The total time to process 30 samples with a count time of 30 minutes was 20 hours.

5.2 Options for Emergency Screening

In an emergency situation, many approaches to screening for radioactivity in urine are possible. Organizations interested in developing this screening capability should identify the information that would be most useful and then consider the approaches and variations to determine which methods would prove beneficial. Some examples to consider are:

1. ***MDA Consideration***: As described in Part 4, MDA is a function of sample/detector size, count time, and background.

- Sample and detector size were found to be optimum in a 1:3 mix of urine and UGXR™ respectively. This mix reliably yields a useful MDA, with minimal chance of an unacceptably quenched (low efficiency) sample. See Part 4 for more discussion of sample and detector size.

- MDA has been thoroughly researched and addressed for constant count times and the optimum sample/detector size. The urgency associated with emergency screening, however, may require varied count times. Because the critical level is inversely proportional to the square root of the count time, MDA is nearly inversely proportional to the square root of the count time, as shown by Currie:[1]

$$L_c \propto \frac{1}{\sqrt{t}} \Rightarrow MDA \approx\propto \frac{1}{\sqrt{t}}$$

Thus quartering the count time only increases the MDA by roughly a factor of two. Similarly, quadrupling the count time only reduces the MDA by roughly half. This important concept is often overlooked. Investigators should be aware that shorter count times do not diminish results as one might expect; conversely, longer count times do not improve the results as one might expect. SNL used a sample of typical urine, a 5-ml aliquot plus 15 ml UGXR™, to illustrate this concept. Figure 19a shows the count times and resulting MDAs for gross alpha screening, although the general shape of the relationship would apply to any radioactivity measurement, as shown in Figure 19b.

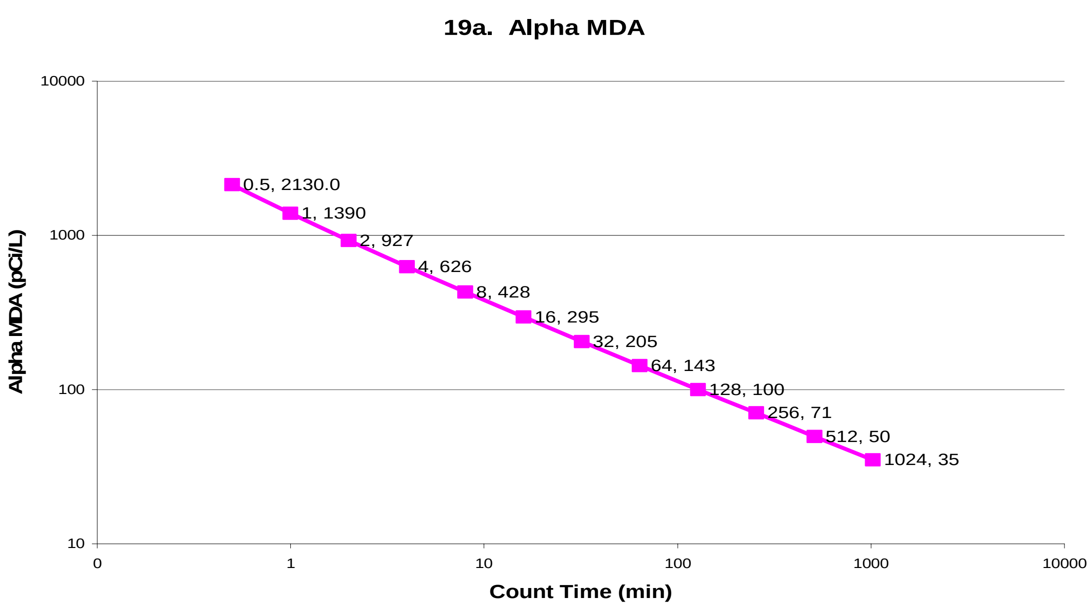

Figure 19. (continued on the next page)

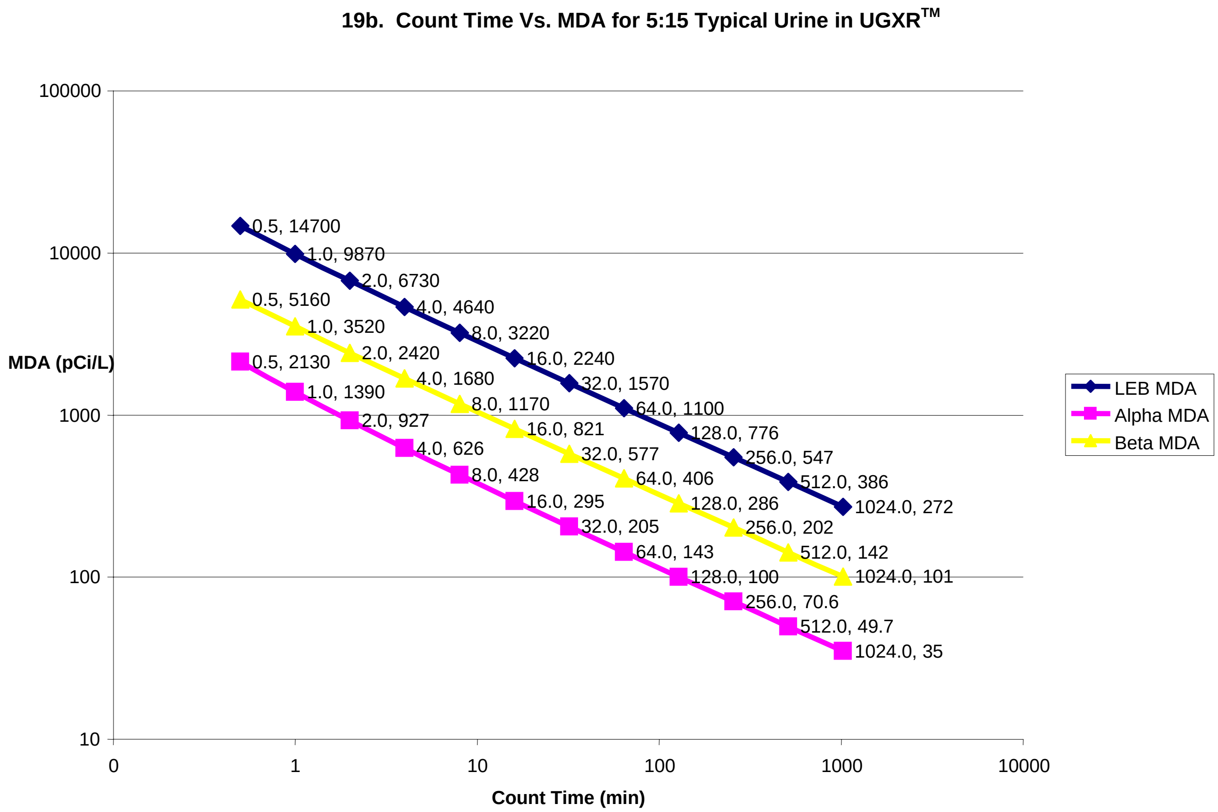

Figure 19. Count time vs. MDA

These figures shows single samples counted repeatedly, with all parameters held constant except count duration. Note the logarithmic scale of the axes. With many samples to analyze, counting the large batches for short times, to single out higher activity or otherwise interesting samples for further investigation, can be an effective approach.

- Sample background is typically an uncontrollable consideration, except in sample size. As explained above background is a minor factor compared to others when determining MDA and optimum sample size.

2. ***Appearance of Urine Samples***: RPSD hypothesized that an experienced technician who has processed many routine samples could judge the quench value that would result when mixed with scintillating cocktail solely by the urine's appearance (clarity, color, consistency). This knowledge would be useful in a screening process to judge the proper sample volume to use, perhaps improving the speed of the screening process. The hypothesis was tested and results are recorded in Table 5.

Table 5. Quench Estimation

Sample Number	Tech #1 Estimate	Tech #2 Estimate	Measured Quench
1	250	230	**249**
2	195	160	**141**
3	190	170	**169**
4	190	170	**155**
5	220	230	**219**
6	250	220	**219**
7	200	175	**168**
8	185	135	**142**
9	270	250	**242**

As Table 5 shows, an experienced technician at times can judge the quench by the urine's appearance, but such educated guesses are not reliably accurate to any useful degree. Further discussion is not necessary because for approximately 99% of urine samples, a 5:15 mix of urine and UGXR^{TM} results in an acceptable quench value. (See Part 4.5 for further discussion.)

3. ***Real-Time Analysis:*** The Tricarb 2500TR and similar LSC machines used by RPSD offer a Spectraview^{TM} function, which allows real-time viewing of the radiation spectrum and raw count data as it is being collected. In an urgent or emergency situation, an experienced technician can make a nearly immediate judgment of each sample as it is counted. There are several advantages to this option:

- Gross judgments with regard to the amount of radioactivity present in the urine can be made; i.e., the question "Is it a lot or is it a little?" can be asked and answered.

- A subjective judgment can be made when enough information has been collected about a sample. This may help improve the processing time of the samples through the screening process.

- The Spectraview^{TM} information can convey characteristics of the radioactivity, such as whether it is a beta or alpha emitter or a mixture. For instance, Figure 20 shows the spectrum of alpha emitter Cm-244 (and its daughter). It is fairly sharp, because alphas are monoenergetic. The beta spectrum indicates that counts assigned as such are actually spillover from alpha to beta channels due to the humped shape rather than a smeared shape characteristic of actual beta detections.

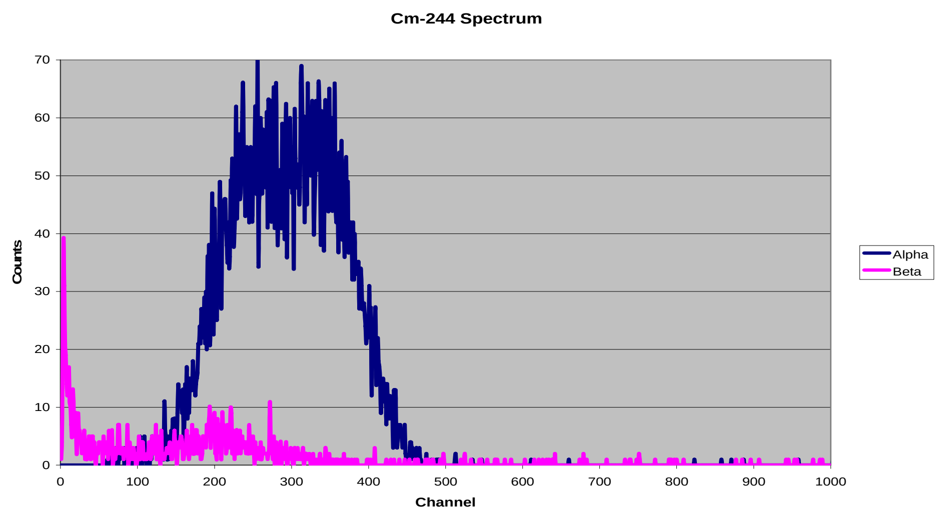

Figure 20. Alpha to beta crosstalk

- Figure 21 shows a beta spectrum. It is more smeared than the spectrum shown in Figure 20 because unlike alphas, betas are not monoenergetic (due to concurrent neutrino emission). The alpha spectrum is also smeared (not monoenergetic) and therefore indicates crosstalk from beta to alpha channels.

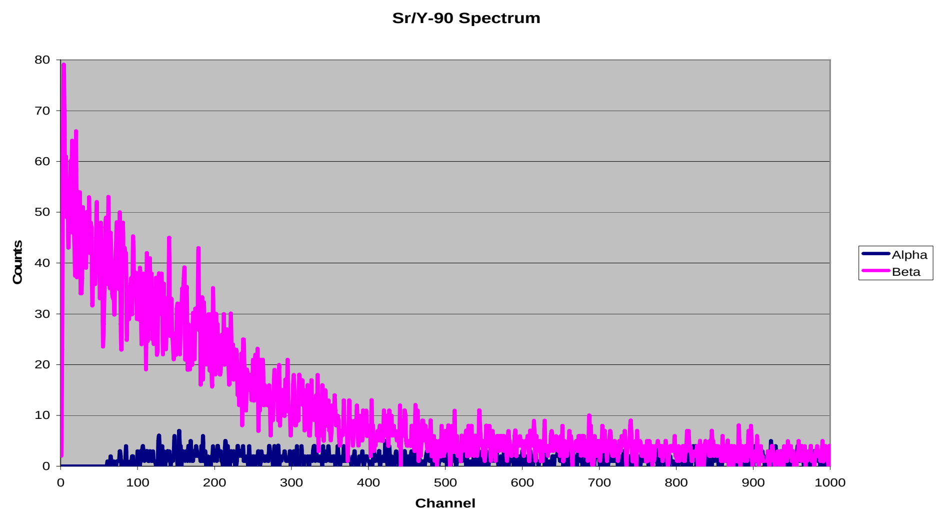

Figure 21. Beta to alpha crosstalk

- Figure 22 shows a mixed spectrum of alpha and beta emitters. The monoenergies of the alpha emitters are evident, as is the smeared nature of the beta emitters.

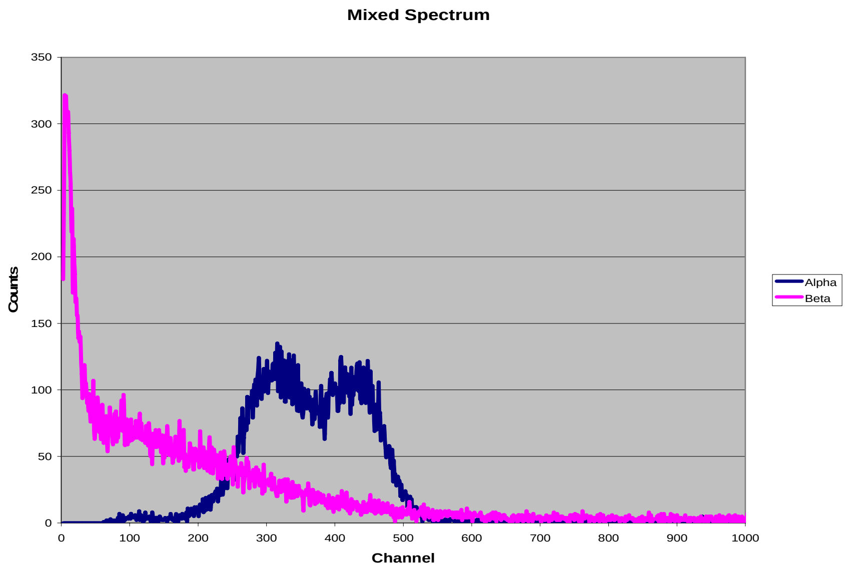

Figure 22. Mixed alpha and beta spectrum

4. ***Statistical Options:*** The Tricarb 2500TR and similar LSC machines used by RPSD are capable of applying statistical tests to each sample. These tests are *weak sample rejection* and *strong sample acceptance*. In the weak sample rejection test, if the sample is found not to contain significant radioactivity (less than a certain number of counts occur within a preset time interval), the sample is rejected and the next sample in the queue is analyzed. In the strong sample acceptance test, a count error percentage is specified at which it is deemed that data are certain enough to move on to the next sample. In an urgent or emergency situation, these tests can shorten the count time of a batch.

6. CONCLUSIONS

Should a population receive an uptake, valuable information could be obtained quickly by screening the urine from a sample of the population and analyzing the urine using liquid scintillation techniques. For a sample-to-cocktail ratio of 1:3, a typical range of discriminator settings was found to be 104-115. Using a mid-range discriminator setting is adequate to perform screening in an emergency or other situation when the nuclide or mix of nuclides is not known. This will typically yield approximate results correct to within a factor of two.

Efficiency curves can be generated for both alphas and betas simultaneously by producing added spikes of a single value and adding a quenching agent in varying amounts to the scintillation cocktail. The efficiency curve is generated using a least squares fit by a convenient natural empirical equation:

$$\varepsilon = \varepsilon_0 - e^{-P1*TSIE^{P2}}$$

where

ε_0 = the efficiency found in an unquenched spike.

P1 and *P2* = fitting parameters.

MDA is also a consideration when determining the optimum sample size. Considering detector/sample combinations constant at 20 ml, at 9 ml typical urine failed to combine with the LSC cocktail. The MDA remained relatively constant for urine volumes between 3 and 7 ml. SNL recommends a mid-volume value of 5 ml for screening purposes. The volume will also be dependent on the TSIE value. The volume should be adjusted to achieve a TSIE value within the range of 120–280. This will leave only 1% of the samples unacceptably quenched.

Another factor to consider once the volume is selected is the count time. With many samples to analyze, counting the large batches for short times to single out higher activity or otherwise interesting samples for further investigation, can be an effective approach. Using the suggested guidelines an alpha MDA of approximately 900 pCi/L for a two-minute count time can be achieved. The statistical capabilities of the instrument, such as weak sample rejection and/or strong sample acceptance, can also be used to decrease processing time. In the weak sample rejection test, if the sample is found not to contain significant radioactivity (less than a certain number of counts occuring within a preset time interval), the sample is rejected and the next sample in the queue is analyzed. In the strong sample acceptance test, a count error percentage is specified at which it is deemed that data are certain enough to move on to the next sample.

These guidelines are intended to help users balance the speed of obtaining useful information against such other considerations as the detection limit, sample volume, and count times.

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7. REFERENCES

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2. NUREG/CR-4007. "Lower Limit of Detection: Definition and Elaboration of a Proposed Position for Radiological Effluent and Environmental Measurements." Washington, D.C.: Nuclear Regulatory Commission, 1984.

3. “LB5100-W Low-Background System User’s Guide.” Oxford Instruments, Oak Ridge, TN, 1993.

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APPENDIX A: RADIOACTIVITY IN URINE BY LIQUID SCINTILLATION

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Radioactivity in Urine by Liquid Scintillation

Robert P. Reese
Sandia National Laboratories
For The Centers for Disease Control and Prevention

Minimum Detectable Activity

- Considering variables that affect MDA, which is generally desired to be low:

$$MDA \propto \frac{\sqrt{N_b}}{t\varepsilon V}$$

  - Low background ➔ low MDA.
  - Long counting time ➔ low MDA.
  - High efficiency ➔ low MDA.
  - Large sample volume ➔ low MDA.

Minimum Detectable Activity

- But in liquid scintillation counting (LSC), samples are usually mixed directly with the detector.

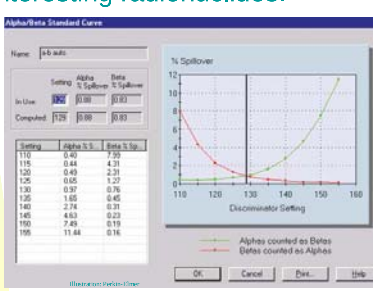
Minimum Detectable Activity

$$MDA \propto \frac{\sqrt{N_b}}{t\varepsilon V}$$

- A larger detector (the scintillating cocktail) is more efficient, but a larger sample volume is desired.
- But larger samples are laden with quenching agents and background noise.
- The total volume of sample plus detector is constant, so a compromise must be found.

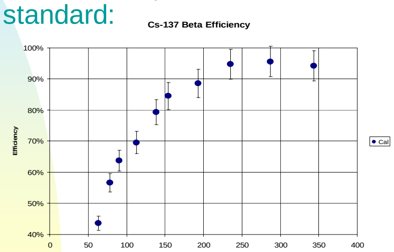
Efficiency Calibration

- First determine discriminator setting for interesting radionuclides.



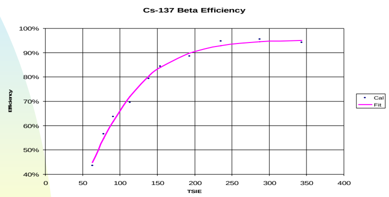
Efficiency Calibration

- Prepare quenched spikes and measure response to external standard:



Efficiency Calibration

- Fit a convenient equation to data:



$$\varepsilon = \varepsilon_0 - e^{-P1*TSIE^{P2}}$$

Where $_0$ = the efficiency found in an unquenched spike.
P1 and *P2* are fitting parameters.

Minimum Detectable Activity

- MDA calculated with simplifications:

$$MDA \approx \frac{2^{3/2} k \sqrt{N_b}}{t\varepsilon V}$$

- In LSC, sample volume + detector volume is constant.
- So larger sample volume sacrifices efficiency and vice versa. It also adds background (though a minor root relationship).

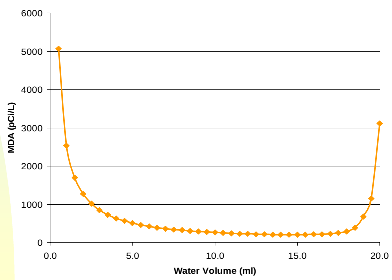
Minimum Detectable Activity

- Empirical observation for a middle-energy beta emitter (Cs-137) and de-ionized water as sample volume and UGXR as detector volume. Total volume 20 ml:

$$MDA \approx \frac{2^{3/2} k \sqrt{N_b}}{t\varepsilon V}$$

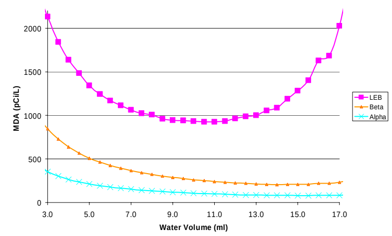
$$\varepsilon = 0.95 - e^{-0.0032*TSIE^{1.3}}$$

t= 1800 s
N_b=1190



Minimum Detectable Activity

- Same general shape seen for alpha emitters, beta emitters and low-energy beta emitters: A flatness or "buffering" near where sample volume and detector volume are equal.



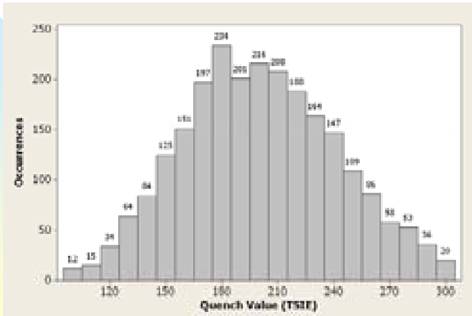
Minimum Detectable Activity

- Repeat MDA experiment with urine.
- But urine varies widely, for many reasons.
- Need to define a typical urine sample.

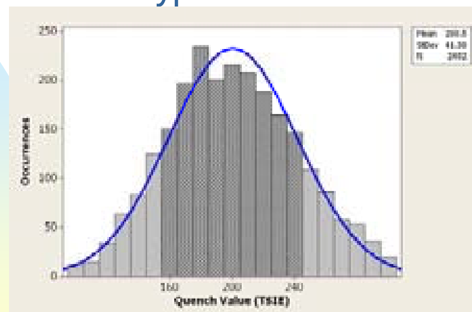
Typical Urine

- As part of SNL internal dosimetry program, urine is analyzed for radioactive constituents (or lack thereof).
- Quench of urine/cocktail 5:15 ml combination has been measured in many samples over several years.

Typical Urine



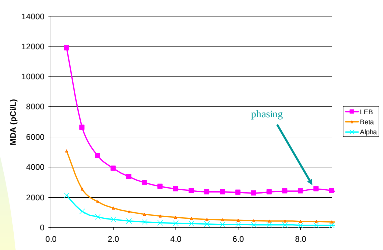
Typical Urine



Normal distribution suggests *typical* urine results in TSIE of 200±40.

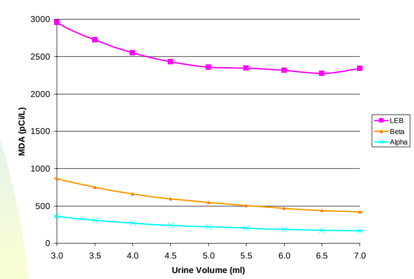
Minimum Detectable Activity

- Repeat MDA experiment with typical urine:



- Same general shape as water at low sample volume.
- Urine "phased" at 9 ml.
- Buffering present, flattest at 3 to 7 ml.

Minimum Detectable Activity



- Zoom on flatness.
- Buffering midpoint is 5 ml.

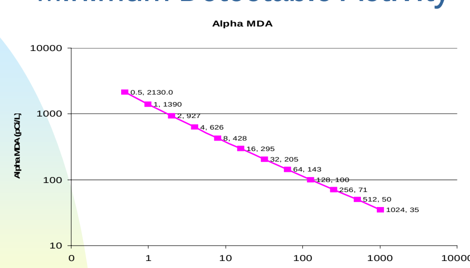
Minimum Detectable Activity

- MDA calculated with simplifications:

$$MDA \approx \frac{2^{3/2} k \sqrt{N_b}}{t \quad \varepsilon V}$$

- In any radioactivity measurement, MDA is larger in a "noisy" background. But it is a root relationship.
- This is why there is only a root return on studying a sample longer.

Minimum Detectable Activity



- Typical urine in UGXR at 5:15 ml ratio.
- Single sample counted for different times.
- Note logarithmic scale.

Discuss Urine Screening Considerations

- Low MDA is desirable.
- 5:15 ml urine/UGXR almost always results in acceptable quench.
- Middle level discriminator setting.
- Spectrum analysis.
- Statistical testing.
- Prep time.

Conclusions

- Mid range discriminator 104-115
- Optimum ratio 5:15 (1:3) urine to UGXR
- Count times a balance of speed vs. MDA are not 1 to 1.

APPENDIX B: BASICS OF LIQUID SCINTILLATION

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Basics of Liquid Scintillation

Robert P. Reese
Sandia National Laboratories
For The Centers for Disease Control and Prevention

Liquid Scintillation Counting (LSC)

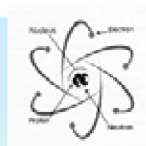
Its purpose:

- Quickly detect certain types of radioactivity.
- Gain rudimentary information about the radioactivity.

What is Liquid Scintillation Counting (LSC)?

Answer this by answering these questions:

- What is radioactivity?
- How does charged radiation interact with other matter?
- What is scintillation?
- What is a liquid scintillating cocktail?



Review of Radioactivity

- Familiar matter is made up of atoms.
- Atoms are made of nuclei and shell electrons.
- Nuclei are made of various combinations of protons and neutrons.
- So atoms are made of various combinations of electrons, protons and neutrons.

Review of Radioactivity

- Electrons are slightly massive (1/1800 u) objects of single negative charge.
- Protons are massive objects (1 u) of single positive charge.
- Neutrons are massive objects (1 u) of no charge.
- Protons and neutrons are collectively known as “nucleons.”

Review of Radioactivity

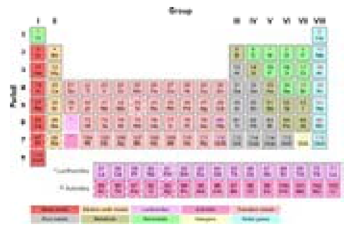
- Positively charged protons in the nucleus repel each other: the *electromagnetic force*.
- Nucleons attract each other: the *strong force* or “nuclear glue.”
- This results in an “intense conflict” between the two forces.

Review of Radioactivity

- Protons add glue *and* charge.
- Neutrons add glue *only*.
- Quantum states fill as nucleons are added, much like shell electrons.
- Some arrangements are stable; most are not.

Chart of the Elements

- Most are already familiar with this chart.



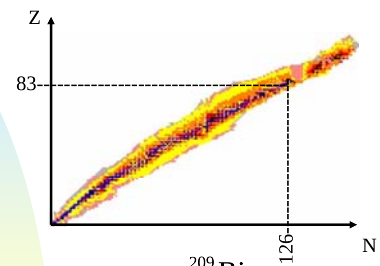
- Logical arrangement of elements based on configuration of shell electrons.
- Gives indications of chemical properties.

Chart of the Nuclides

- Fewer are familiar with this chart.

- Logical plot of significant proton/neutron combinations– the nuclides.
- KAPL/GE/Lockheed wall chart is most common reference.
- Gives indications of nuclear properties.

Chart of the Nuclides

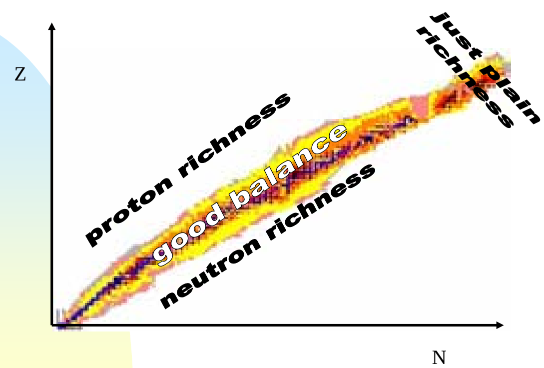


- Example: $^{209}_{83}$Bi
- (There is not a firm convention on the axes.)

Review of Radioactivity

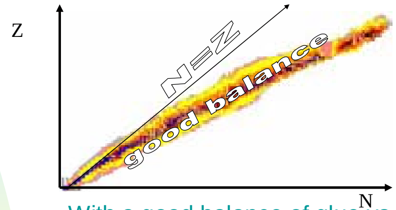
- Four possibilities in these arrangements of nucleons:
  1. Balance of nucleons.
  2. Neutron rich (proton poor).
  3. Proton rich (neutron poor).
  4. Just plain richness (too many protons)– Z>83 guarantees instability.

Chart of the Nuclides



- The four possibilities in arrangements of nucleons

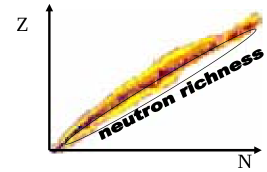
Stability of Nuclei



- With a good balance of glue vs. charge, nuclei tend to be stable.
- As charge accumulates, extra neutron glue is needed. Notice downward concavity of plot.

Review of Radioactivity

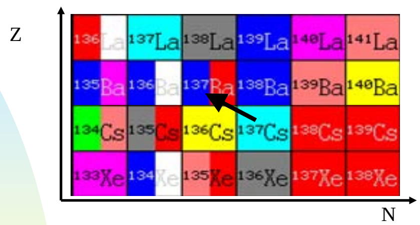
Beta Decay of N-rich nucleus



- Neutrons tend to become protons by flipping out a highly kinetic electron– a beta particle.
- Movement of nucleus' makeup is up and leftward toward stability.

Review of Radioactivity

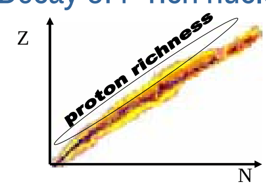
Beta Decay of N-rich nucleus



- Example: $^{137}_{55}\mathrm{Cs} \rightarrow ^{137}_{56}\mathrm{Ba} + \beta^- + \nu$
- Neutrinos are beyond scope of this context, except that they have random momentum, therefore the betas from this reaction will not be of a single energy.

Review of Radioactivity

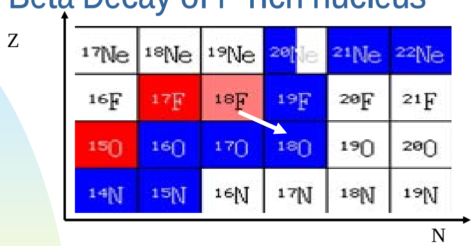
Beta Decay of P-rich nucleus



- Protons tend to become neutrons by flipping out a highly kinetic positron– also a beta particle.
- Movement of nucleus' makeup is right and downward toward stability.

Review of Radioactivity

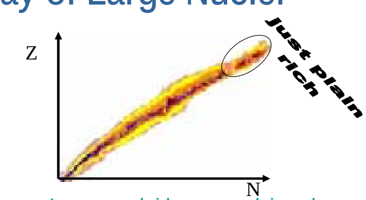
Beta Decay of P-rich nucleus



- Example: $^{18}_{9}\mathrm{F} \rightarrow ^{18}_{8}\mathrm{O} + \beta^+ + \nu$
- The positron is antimatter, soon annihilated (useful for positron emission tomography– PET).

Review of Radioactivity

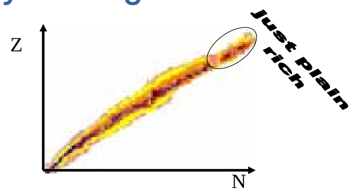
Decay of Large Nuclei



- Large nuclei have repulsive charge forces on par with the attractive strong forces. Also their size is on par with range of the strong forces-- the "glue" can break down.
- Multiple decay processes are possible.
  1. Alpha emission
  2. Beta emission
  3. Fission

Review of Radioactivity

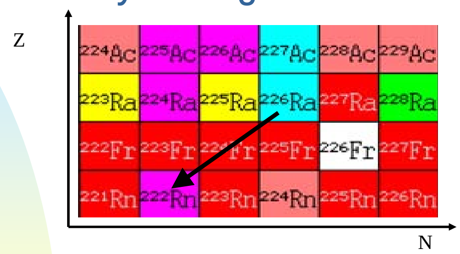
Decay of Large Nuclei



- The alpha particle is a helium nucleus.
- It is two protons plus two neutrons which can "tunnel out" of heavy nuclei.

Review of Radioactivity

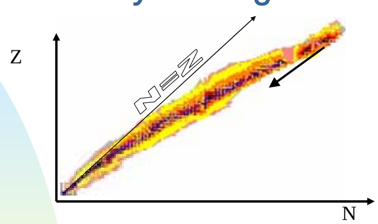
Decay of Large Nuclei



- Alpha decay example: $^{226}_{88}\mathrm{Ra} \rightarrow ^{222}_{86}\mathrm{Rn} + ^{4}_{2}\alpha$
- Movement of the nucleus' makeup is left and downward toward stability.

Review of Radioactivity

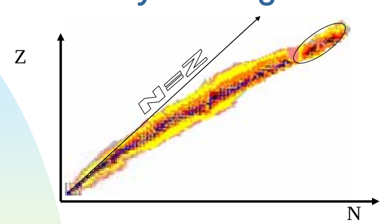
Decay of Large Nuclei



- Movement of the nucleus' state is left and downward with same slope as N=Z.
- Therefore the decay product tends to be N-rich.

Review of Radioactivity

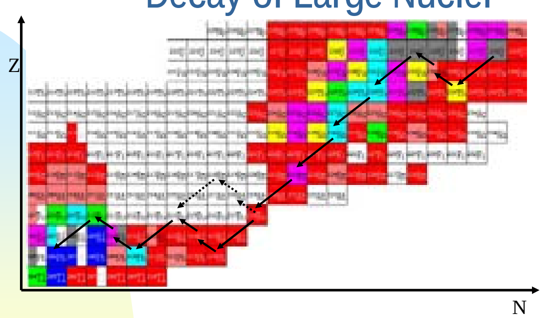
Decay of Large Nuclei



- Large nuclei decay products are *both* large and neutron rich.
- Therefore they tend to decay by *both* alpha and beta methods.
- This leads to zigzagging "decay chains."

Review of Radioactivity

Decay of Large Nuclei

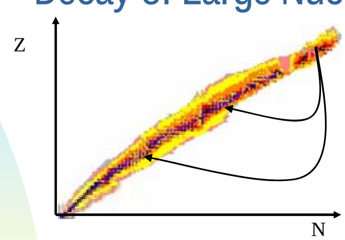


- Example: $^{238}_{92}\mathrm{U} \rightarrow ^{206}_{82}\mathrm{Pb} + 6\beta^- + 8^{4}_{2}\alpha$

(Plus neutrinos)

Review of Radioactivity

Decay of Large Nuclei



- Large nuclei can also split or "fission" into two smaller nuclei.
- Example: $^{235}_{92}\mathrm{U} \rightarrow ^{137}_{55}\mathrm{Cs} + ^{98}_{37}\mathrm{Rb}$
- Fission products tend to be N-rich.

Review of Radioactivity

- As these processes occur, the nucleus left behind will usually “settle” into its new configuration.
- When it does this, additional energy is released in the form of an energetic photon, or “gamma ray.”
- Like agitating a jar of marbles: they settle and a little heat is released.
- Gamma radiation is tangentially pertinent to this LSC discussion.

Review of Radioactivity

- Other decay processes are possible such as:
  1. Nucleon emission.
  2. Electron capture with subsequent gamma and/or shell electron (Auger) emission.
  3. Internal transition (a delayed settling of the nucleus).
- These are *not* pertinent to this particular discussion of LSC.

Liquid Scintillation

Review of Radioactivity

- *Activity* is the rate at which a radioactive substance decays.
- It is a number of events per unit time.
- Units:
  - Metric: the Becquerel (Bq) = 1 decay per second.
  - Traditional: the Curie (Ci) = 3.7×10^{10} decays per second.
- *“Activity”* often refers in practice to activity per some unit of measure of the substance containing the radioactivity (such as volume or mass). This is more precisely called *specific* activity.

Review of Energy

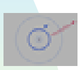
- An object such as an electron with single charge moved (by whatever means) through an electric potential of one volt will have energy of one electron-volt or eV.
- An eV is small compared to everyday energy visualizations, but is useful on the atomic and nuclear scales of energy.

Review of Energy

Electron-Volt in Perspective

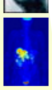
Some non-nuclear energies:

- Sodium lamps emit light due to movement of outer shell electrons at about 2 eV.



- Removing the electron from a hydrogen atom (ionization) requires about 14 eV.

- Tungsten inner shell electron excitement produces X-rays with energy 70,000 eV (70 keV).



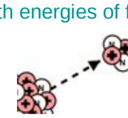
- Positron emission tomography (PET): The “P” in PET has energy of 511 keV (antimatter-matter annihilation).

Review of Energy

Electron-Volt in Perspective

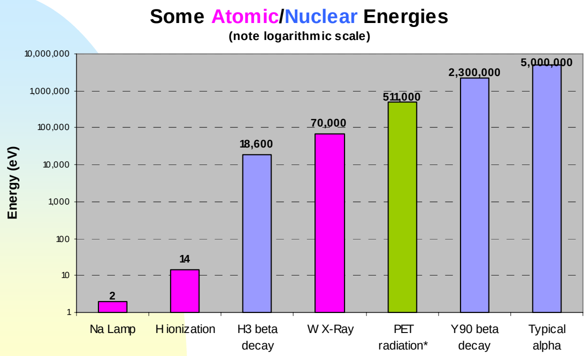
Some nuclear energies:

- Having overcome the intense conflict, alpha and beta particles leave the parent nucleus with high energy.
- Betas with energies one to thousands of keV.
- Alphas with energies of four to six MeV.



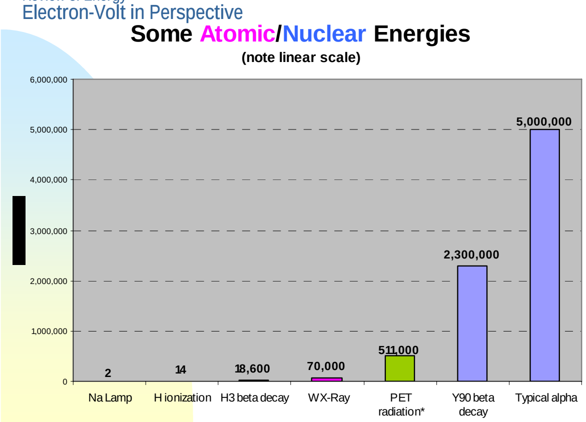
Review of Energy

Electron-Volt in Perspective



Review of Energy

Electron-Volt in Perspective



Review of Energy

- Chemical reactions have unique energies (Q-values) depending on the configuration of new atomic arrangements.
- Just like with chemical reactions, the change due to a nuclear reaction (i.e. decay) has a unique Q-value.

Review of Energy

Nuclear* Q-Value Examples

$^{137}_{55}\mathrm{Cs} \rightarrow ^{137}_{56}\mathrm{Ba} + \beta^- + \nu$ Q=514 KeV

$^{18}_{9}\mathrm{F} \rightarrow ^{18}_{8}\mathrm{O} + \beta^+ + \nu$ Q=634 KeV

$\beta^+ + \beta^- \rightarrow 2\gamma$ Q=1.02 MeV (511 KeV x 2)*

$^{226}_{88}\mathrm{Ra} \rightarrow ^{222}_{86}\mathrm{Rn} + ^{4}_{2}\alpha$ Q=4.78 MeV

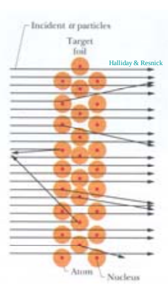
$^{238}_{92}\mathrm{U} \rightarrow ^{206}_{82}\mathrm{Pb} + 6\beta^- + 8^{4}_{2}\alpha$ Q=50 MeV

$^{235}_{92}\mathrm{U} \rightarrow ^{137}_{55}\mathrm{Cs} + ^{98}_{37}\mathrm{Rb}$ Q=190 MeV

*Annihilation is not generally considered nuclear.

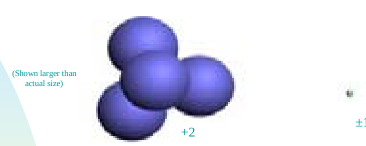
Charged Particles Interact

- A charged energetic particle will interact as it passes through matter by electromagnetic collisions. The material will be excited and disturbed.
- Analogy: a car which *careens* off a highway will bounce off of the trees in a forest. Branches will break and roots will be upped.



Famous Rutherford Experiment

Charged Particles Interact

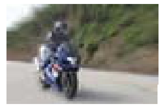


- Betas have mass 1/7000 that of alphas but only half the charge. (Charge is a minor consideration, except that charge *is* present.)
- Alphas can have energy thousands that of betas but only twice the charge. (Again, charge is a minor consideration.)

Charged Particles Interact

Analogy (a stretch)

Dump Truck

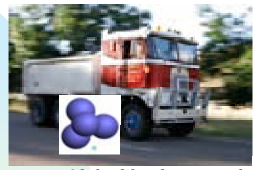


Motorcycle

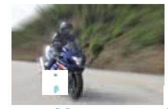
- A dump truck moving at 20 miles per hour has much energy– of a massive moseying type.
- A motorcycle moving at 200 miles per hour also has much energy– of a light speeding type.
- (Classically, $E = \frac{1}{2} m v^2$)

Charged Particles Interact

Analogy



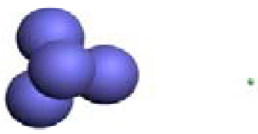
Alpha like dump truck



Beta like motorcycle

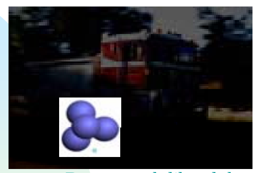
- When the dump truck *careens* into the woods, it will devastate (interact), but stop soon, interacting in a short time and distance.
- When the motorcycle *careens* into the woods, it will interact and stop eventually in a longer time and distance. (Apologies to Bill Watterson.)

Charged Particles Interact

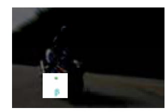


- Being charged energetic particles, the interactions will be excitations and ionizations of the forests of matter through which they travel.
- The alpha will produce many ionizations and excitations in a short time and distance.
- The beta will produce many ionizations and excitations in a long time and distance.

Charged Particles Interact



Dump truck like alpha



Motorcycle like beta

- Keep these concepts of similarities and differences in mind as we move to the next subject:
- Scintillation➔

Cliff Claven Moment

What is Scintillation?

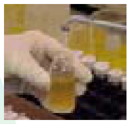
- From the Latin word for sparks: *scintillata.*
- In this context, a *scintillation* is flash of light emission in response to ionization and excitation by radioactivity of the scintillating medium.
- It's a conversion of keV or MeV in energy to light pulses of eV.
- Fluorescence and phosphorescence chemically enable scintillation.

What is Scintillation?

Many Compounds Scintillate

- Inorganic crystals such as sodium iodide (NaI) used in isotope imaging. (High density good for gamma detection.)
- Zinc sulfide, used by Rutherford for alpha detection.
- Organic solids such as scintillating polymers.
- Organic liquids such as toluene/fluor *cocktails* good for alpha and beta detection: *Liquid scintillation counting* (LSC).

Liquid Scintillation Counting (LSC)

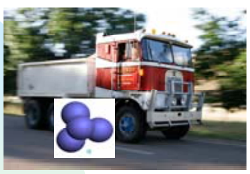


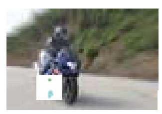
- Liquid samples are often mixed with scintillation cocktails to measure radioactivity in the sample.
- This places the sample in direct contact with the detector, making it possible to detect lower energy particles such as the 18.6 KeV tritium beta.
- Sandia uses a non-toxic biodegradable cocktail: Ultima Gold Extended Range (UGXR).

Liquid Scintillation

- The conversion of up to MeV of energy to light pulses of eV implies that higher energy results in more intensity of the pulses➔ The light pulse is proportional to the input excitation.

Liquid Scintillation

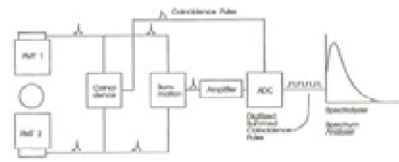




- Light pulses from alphas and betas are different due to the way they react and their differences in energy, like the differences in damage to forests as dump trucks and motorcycles *careen* off the road.
- Alpha scintillations will be bright and short-lived.
- Beta scintillations will be dim and long-lived.
- This enables differentiation or *discrimination* between alphas and betas in LSC (with the right equipment).

Liquid Scintillation

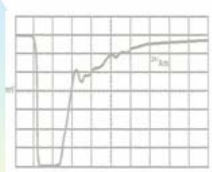
Discrimination

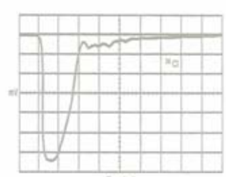


- Scintillations are collected by photo-multiplier tubes and converted to electronic signals of *time profile* and *amplitude* indicating the nature of the original radiation.
- This is time-resolved pulse decay analysis or TRPDA.

Liquid Scintillation

Discrimination





- On the left, a TRPDA of an alpha emitter.
- On the right, a TRPDA of a beta emitter.

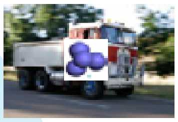
Liquid Scintillation

Discrimination

- Discrimination is not perfect: Occasionally a beta will be mistaken for an alpha and vice-versa (known as "overlap", "crosstalk", "spillover").
- We must "teach" the electronics to minimize the mistakes by adjusting the electronic discriminating component– the discriminator.

Liquid Scintillation

Discrimination



- In one extreme, the discriminator will recognize all decays as alphas.

- In the other extreme, the discriminator will recognize all decays as betas.

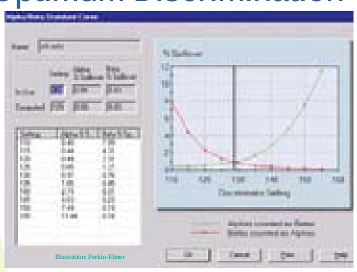
Liquid Scintillation

Discrimination

- The happy medium is the minimization of mistakes.

Liquid Scintillation

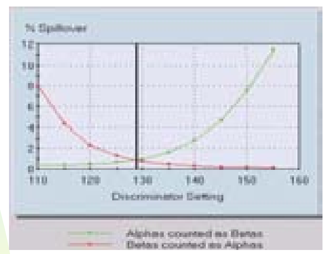
Optimum Discrimination



- Optimum discrimination minimizes the misinterpretation of alphas as betas and vice versa.

Liquid Scintillation

Optimum Discrimination



- The discriminator in this case is electronically adjusted on an arbitrary scale (0-256).

Liquid Scintillation

Optimum Discrimination

- Optimum discrimination depends some on the energies of the alphas and betas being detected.
- Energies of the particles depend on the reaction Q-value, unique to the radionuclide.
- So different combinations of alphas and betas will result in different optimum discrimination settings.

Liquid Scintillation

Optimum Discrimination

- Sandia studied 15 different alpha and beta combinations, three alphas and five betas:

Alpha Emitters		Beta Emitters	
Nuclide	Energy (KeV)	Nuclide	Max Energy (KeV)
Th-230	4700	H-3	19
Pu-239	5150	Ni-63	67
Cm-244	5800	Cs-137	514
		Cl-36	710
		Sr/Y-90*	2190

*2190 KeV beta is from Y-90.

Optimum Discrimination

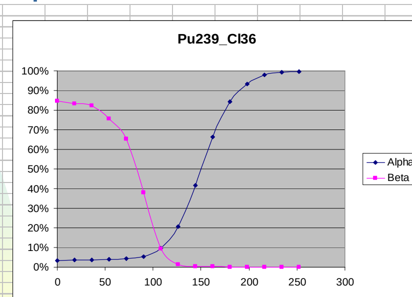
Discriminator Settings

Beta Emitters		Alpha Emitters: Th-230	Pu-239	Cm-244	Maximum Beta Energy (KeV)
	H-3	90	92	93	19
	Ni-63	45	41	69	67
	Cs-137	104	105	108	514
	Cl-36	106	108	110	710
	Sr/Y-90	111	112	115	2190
	Alpha Energy (KeV)	4700	5150	5800	

- Result, a range of optimum discriminator settings.
- But for the larger beta energies and a wide range of alpha energies, the optimum discriminator settings fall within a narrow range.

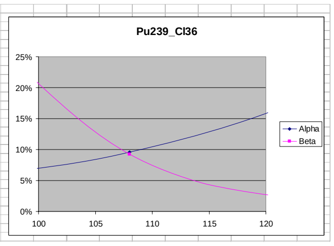
Optimum Discrimination

	% Spillover	
Setting	Alpha	Beta
0	3.3%	84.6%
18	3.8%	83.3%
36	3.8%	82.3%
54	3.9%	75.7%
72	4.5%	65.2%
90	5.3%	37.9%
108	9.6%	9.2%
126	20.5%	1.3%
144	41.6%	0.5%
162	66.3%	0.2%
180	84.5%	0.1%
198	93.4%	0.1%
216	97.9%	0.1%
234	99.3%	0.0%
252	99.6%	0.0%
Optimum:		
108	9.59%	9.21%



Optimum Discrimination

	% Spillover	
Setting	Alpha	Beta
0	3.3%	84.6%
18	3.8%	83.3%
36	3.8%	82.3%
54	3.9%	75.7%
72	4.5%	65.2%
90	5.3%	37.9%
108	9.6%	9.2%
126	20.5%	1.3%
144	41.6%	0.5%
162	66.3%	0.2%
180	84.5%	0.1%
198	93.4%	0.1%
216	97.9%	0.1%
234	99.3%	0.0%
252	99.6%	0.0%
Optimum:		
108	9.59%	9.21%



- This implies that for gross screening, a mid-level setting is a good compromise when nuclides are unknown or mixed, (such as fission products).

Minimum Detectable Activity

- Again, *activity* is the rate at which a radioactive substance decays.
- *Minimum detectable activity* (MDA) is the activity that, if present in a substance, we have a pre-determined likelihood of detecting.
- The key to measuring MDA is in how we come to this pre-determination.

Minimum Detectable Activity

- For our purposes, we adopt a simplified version of L.A. Currie's method:

$$MDA \approx \frac{2L_c}{\varepsilon V}$$

Where L_c is the "critical limit."
is counting efficiency.
V is volume (or mass m or area A to convert to *specific* activity).

Critical Limit

- L_c is the highest value of activity reportable by a counter for a sample with no activity (to a specified confidence).
- The specified confidence is usually set to 95%.
- In other words, 5% of the time, a sample with no activity will be reported above the critical limit.

Critical Limit

- Mathematically:

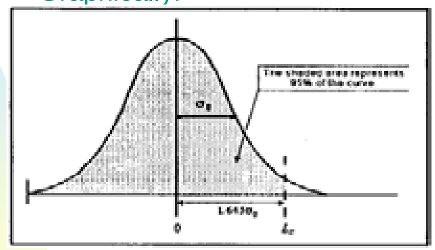
$$L_c = k\sigma_0$$

Where *k* is a multiplier that for Gaussian distributions of random events specifies the degree of confidence. (For 95% confidence, *k*=1.645.)

And ${}_0$ is the standard deviation (error) of the Gaussian distribution of a sample with no activity.

Critical Limit

- Graphically:



In other words, 95 % of samples with no activity will be reported below the critical limit.

Error: Standard Deviation

- Mathematically:

$$\sigma_0 = \frac{\sqrt{2N_b}}{t}$$

Where N_b is the number of background counts or “noise” detected in a time *t.*

This is treated in general in statistics courses and in detail in L.A. Currie’s paper.

- Key: Error increases with background noise, and decreases with analysis time.

Detector Efficiency

- Again, *Activity* is the rate at which a radioactive substance decays.
- All real detectors only detect a portion of the decays.

Detector Efficiency

- Of the decays that occur, only a fraction can be detected. That fraction is the *efficiency* of a detection system.

$$\varepsilon = \frac{n_d}{n}$$

Where n_d is the number of detections of *n* events.

- Analogy: Of the light emitted from a light bulb, only a small percentage enters your eyes and is detected.

LSC Efficiency

- Found by measuring UGXR/Sample mix response to an external gamma source. The *external standard.*
- The gamma source causes Compton excitation and ionization in the mixture.

Efficiency

LSC Efficiency

- UGXR is the detector; PMT, MCA, computer just process information supplied by UGXR/Sample.

Efficiency

LSC Efficiency

- But we need an *index* for the response to the external standard.
- So we *transform* the spectral response to the external standard into an index.

LSC Efficiency

TSIE

- TSIE is the acronym for the **T**ransformed **S**pectral **I**ndex the **E**xternal standard.
- It is a measure of *quench*, or the interfering effect of chemicals and colorings and dirt, etc. preventing scintillations (light) from being passed to the instrumentation.
- The TSIE scale ranges arbitrarily from 0 to about 400 for UGXR.

LSC Efficiency

TSIE

- We must calibrate this scale for it to be useful in measuring efficiency.
- This is done by adding known amounts of radioactivity to UGXR (internal standards or *spikes).*
- Then we add a quenching agent (nitromethane) in varying amounts and measuring the TSIE.

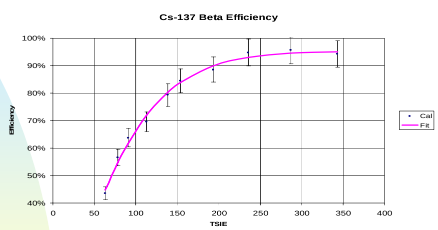
LSC Efficiency

TSIE Calibration

- The more quenched a sample is, the less efficient the detector (and vice versa):
- This is done by adding known amounts of radioactivity to UGXR (internal standards or *spikes).*
- Then we add a quenching agent in varying amounts and measuring the TSIE.

LSC Efficiency

TSIE Calibration Example

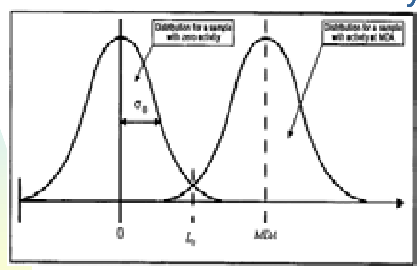


- Low TSIE (high quenching) means low efficiency .

Minimum Detectable Activity

- Again, MDA is the activity that, if present in a substance, we have a pre-determined likelihood of detecting.
- Since L_c is set to 5% false positive, it is logical to set MDA to 5% false negative– a simplifying and customary setting.

Minimum Detectable Activity



Minimum Detectable Activity

- For radiological measurements in general, a low MDA is desirable.
- Combining these concepts, examine the MDA equation:

$$\left.\begin{aligned} MDA &\approx \frac{2L_c}{\varepsilon V} \\ L_c &= k\sigma_0 \\ \sigma_0 &= \frac{\sqrt{2N_b}}{t} \end{aligned}\right\} \quad MDA \approx \frac{2^{3/2}k\sqrt{N_b}}{t\varepsilon V}$$

Minimum Detectable Activity Conclusions

- Removing constants, examine each variable:

$$MDA \propto \frac{\sqrt{N_b}}{t\varepsilon V}$$

  - Low background ➔ low MDA.
  - Long counting time ➔ low MDA.
  - High efficiency ➔ low MDA.
  - Large sample volume ➔ low MDA.

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APPENDIX C: DISCRIMINATOR CALIBRATION GRAPHS FOR VARIOUS COMBINATIONS OF ALPHA- AND BETA-EMITTING RADIONUCLIDES

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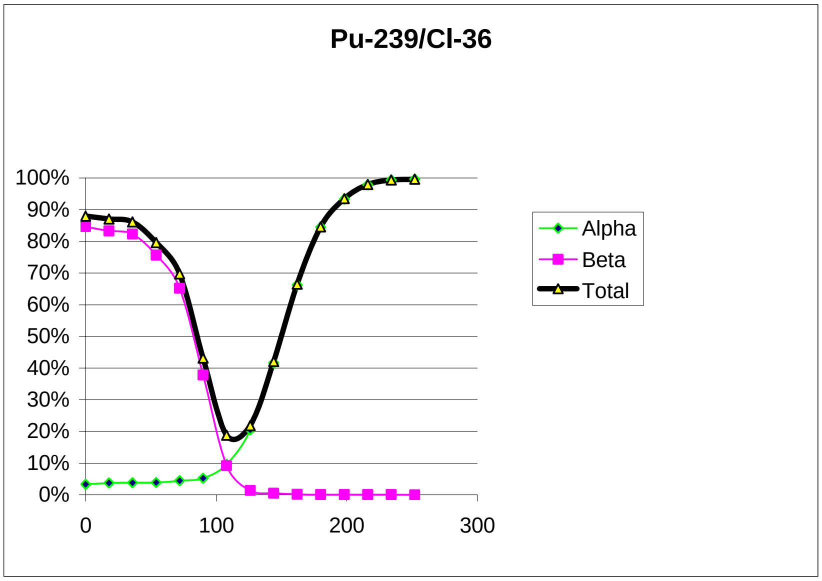
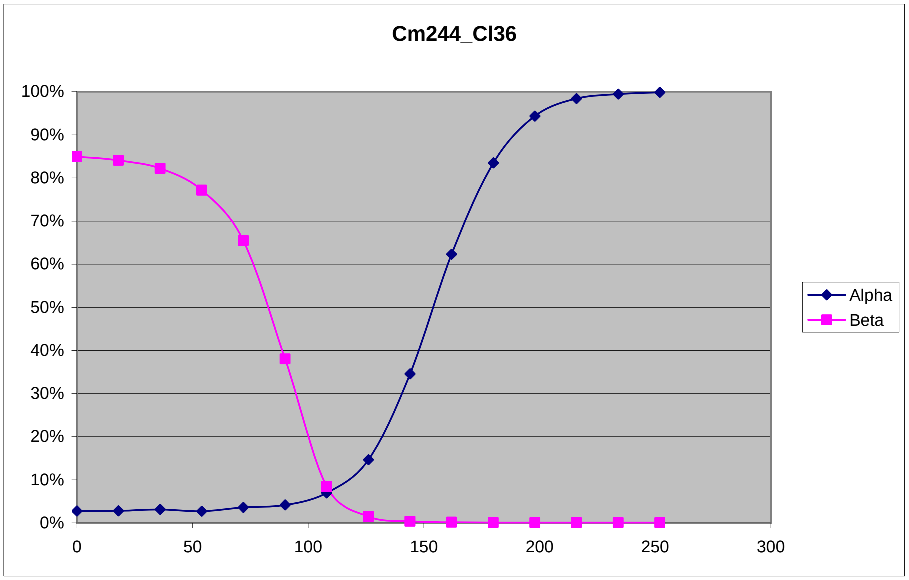
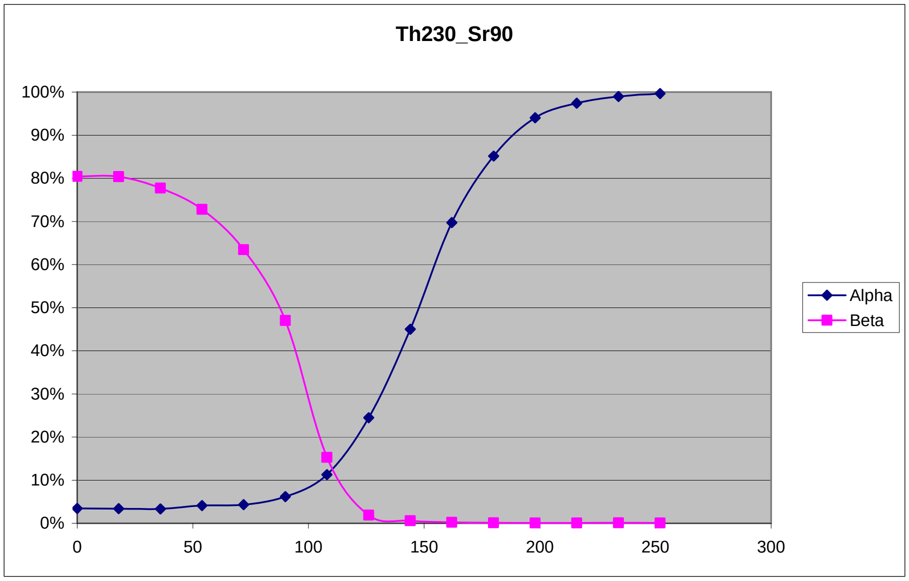
	% Spillover		
Setting	Alpha	Beta	Total
0	3.3%	84.6%	88.0%
18	3.8%	83.3%	87.1%
36	3.8%	82.3%	86.1%
54	3.9%	75.7%	79.6%
72	4.5%	65.2%	69.7%
90	5.3%	37.9%	43.1%
108	9.6%	9.2%	18.8%
126	20.5%	1.3%	21.8%
144	41.6%	0.5%	42.1%
162	66.3%	0.2%	66.5%
180	84.5%	0.1%	84.6%
198	93.4%	0.1%	93.5%
216	97.9%	0.1%	97.9%
234	99.3%	0.0%	99.4%
252	99.6%	0.0%	99.7%
Optimum:			
108	9.59%	9.21%	18.8%



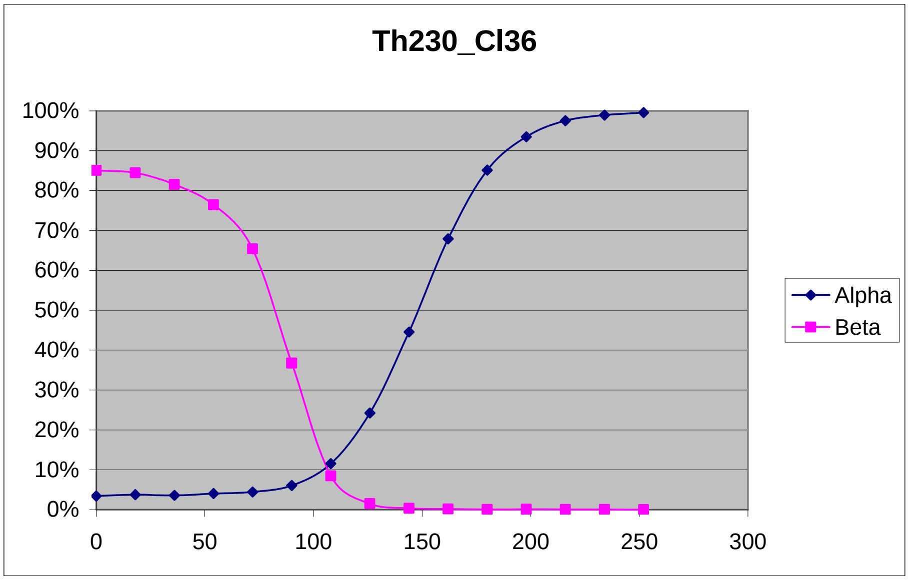
	% Spillover	
Setting	Alpha	Beta
0	2.7%	85.0%
18	2.8%	84.0%
36	3.1%	82.2%
54	2.7%	77.1%
72	3.6%	65.4%
90	4.1%	38.0%
108	7.0%	8.4%
126	14.7%	1.5%
144	34.6%	0.4%
162	62.3%	0.1%
180	83.5%	0.1%
198	94.3%	0.1%
216	98.4%	0.0%
234	99.4%	0.1%
252	99.8%	0.1%
Optimum:		
110	7.39%	6.95%



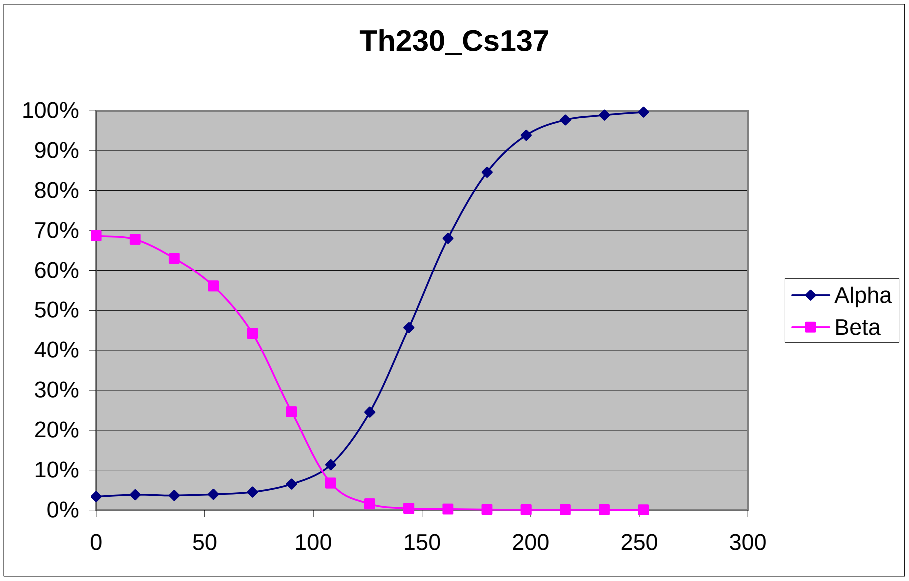
	% Spillover	
Setting	Alpha	Beta
0	3.5%	80.4%
18	3.4%	80.4%
36	3.3%	77.7%
54	4.1%	72.8%
72	4.3%	63.5%
90	6.2%	47.0%
108	11.3%	15.3%
126	24.5%	1.9%
144	45.0%	0.6%
162	69.7%	0.2%
180	85.1%	0.1%
198	94.0%	0.1%
216	97.4%	0.1%
234	99.0%	0.1%
252	99.7%	0.1%
Optimum:		
111	12.92%	11.99%



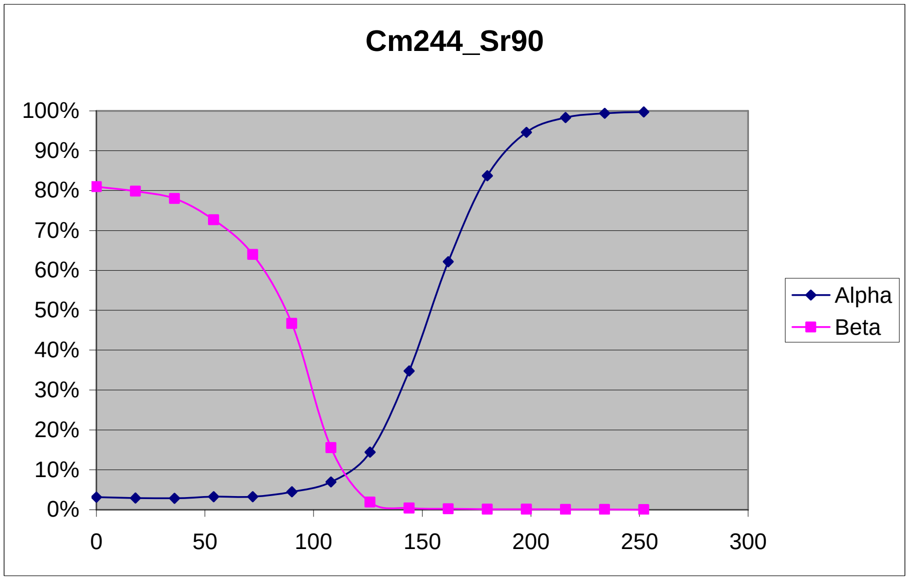
illover
Beta
85.1%
84.5%
81.5%
76.4%
65.4%
36.8%
8.5%
1.5%
0.3%
0.2%
0.1%
0.1%
0.1%
0.1%
0.0%
11.07%



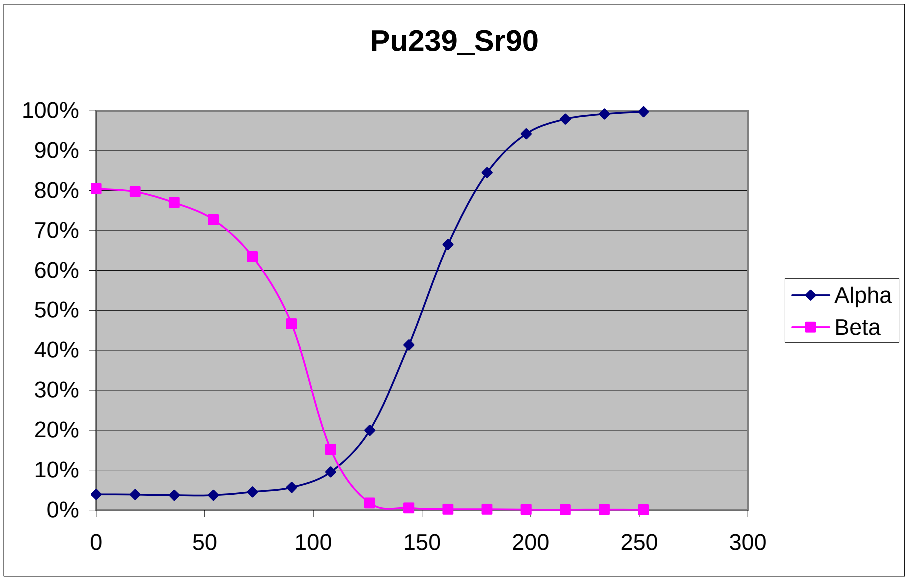
	% Spillover	
Setting	Alpha	Beta
0	3.4%	68.7%
18	3.8%	67.8%
36	3.7%	63.0%
54	3.9%	56.1%
72	4.5%	44.2%
90	6.5%	24.6%
108	11.3%	6.7%
126	24.5%	1.5%
144	45.7%	0.4%
162	68.1%	0.2%
180	84.7%	0.1%
198	93.9%	0.0%
216	97.7%	0.1%
234	99.0%	0.1%
252	99.7%	0.0%
Optimum:		
104	9.77%	10.07%



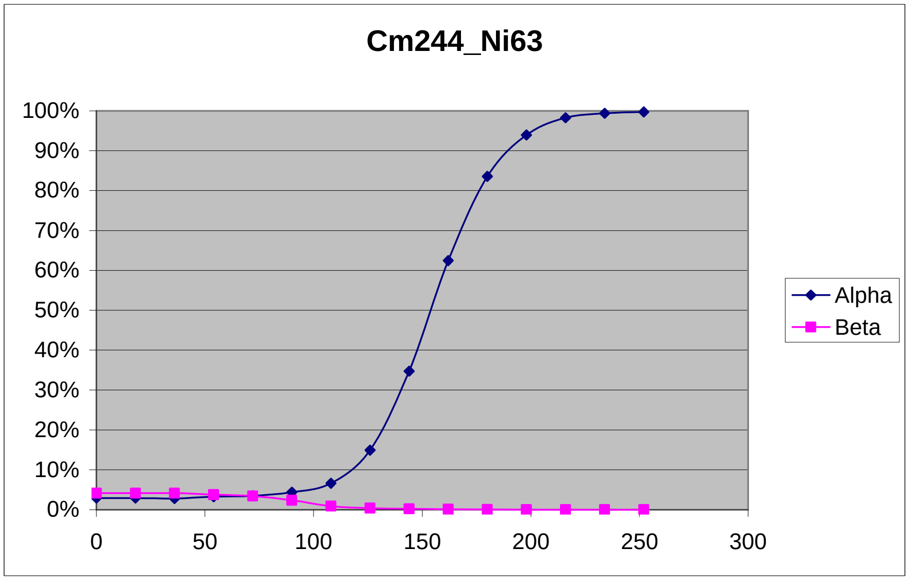
	% Spillover	
Setting	Alpha	Beta
0	3.1%	81.0%
18	2.9%	79.9%
36	2.8%	78.0%
54	3.3%	72.7%
72	3.3%	64.0%
90	4.5%	46.7%
108	7.0%	15.6%
126	14.4%	1.9%
144	34.8%	0.4%
162	62.2%	0.2%
180	83.8%	0.1%
198	94.6%	0.1%
216	98.3%	0.1%
234	99.4%	0.1%
252	99.7%	0.0%
Optimum:		
115	8.80%	8.49%



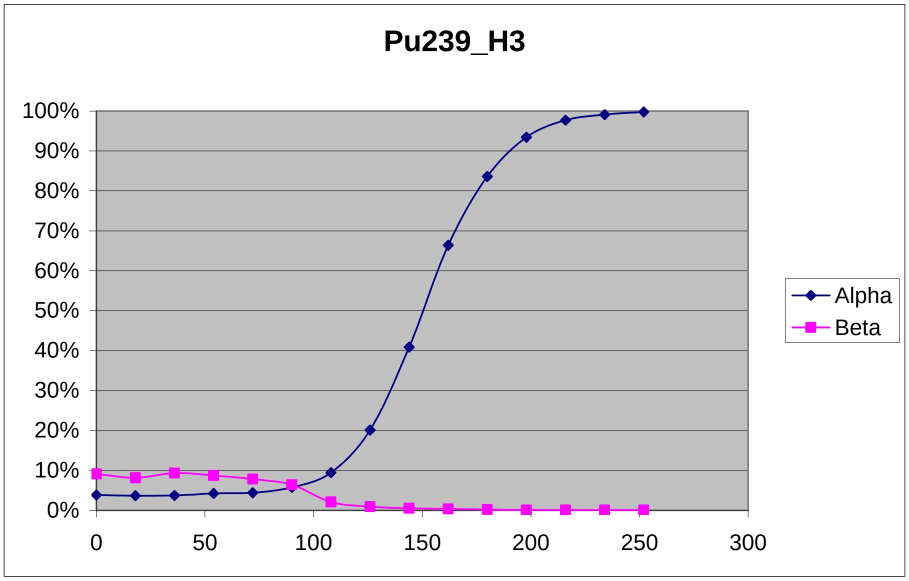
	% Spillover	
Setting	Alpha	Beta
0	4.0%	80.5%
18	3.9%	79.8%
36	3.7%	77.0%
54	3.7%	72.7%
72	4.6%	63.4%
90	5.7%	46.6%
108	9.6%	15.2%
126	20.0%	1.8%
144	41.4%	0.5%
162	66.5%	0.2%
180	84.5%	0.2%
198	94.2%	0.1%
216	97.9%	0.0%
234	99.2%	0.1%
252	99.8%	0.0%
Optimum:		
112	11.11%	10.88%



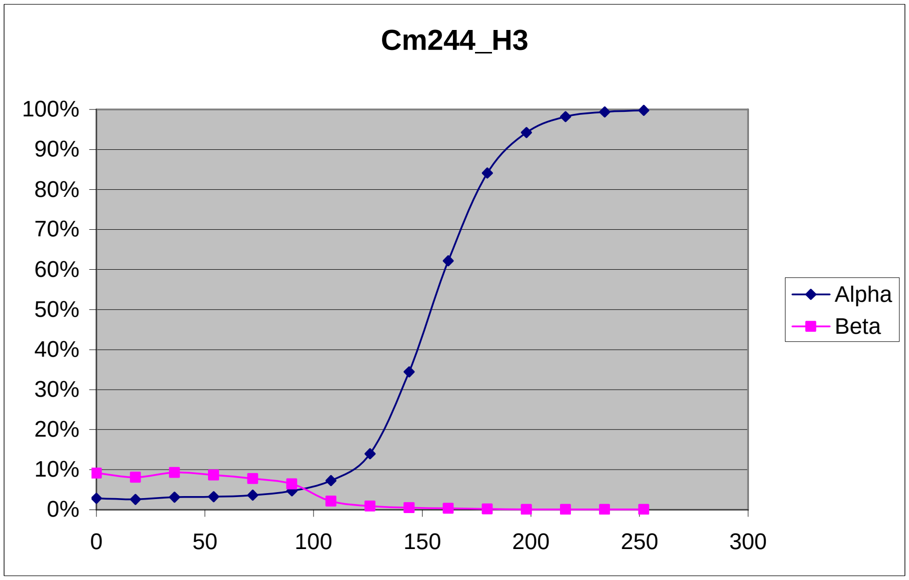
	% Spillover	
Setting	Alpha	Beta
0	2.9%	4.2%
18	2.9%	4.1%
36	2.8%	4.1%
54	3.3%	3.8%
72	3.5%	3.4%
90	4.4%	2.3%
108	6.6%	0.9%
126	14.9%	0.4%
144	34.7%	0.2%
162	62.5%	0.1%
180	83.6%	0.1%
198	94.0%	0.0%
216	98.3%	0.0%
234	99.4%	0.0%
252	99.7%	0.0%
Optimum:		
69	11.11%	10.88%



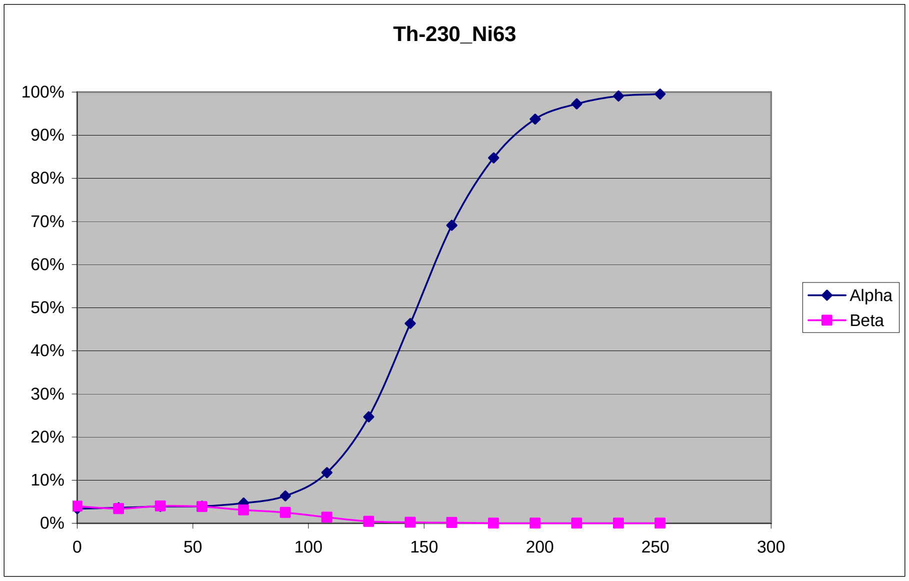
	% Spillover	
Setting	Alpha	Beta
0	3.8%	9.1%
18	3.6%	8.1%
36	3.7%	9.3%
54	4.2%	8.7%
72	4.4%	7.8%
90	5.7%	6.4%
108	9.5%	2.1%
126	20.1%	0.9%
144	40.9%	0.5%
162	66.4%	0.3%
180	83.6%	0.2%
198	93.4%	0.1%
216	97.7%	0.1%
234	99.1%	0.1%
252	99.8%	0.0%
Optimum:		
92	5.89%	5.93%



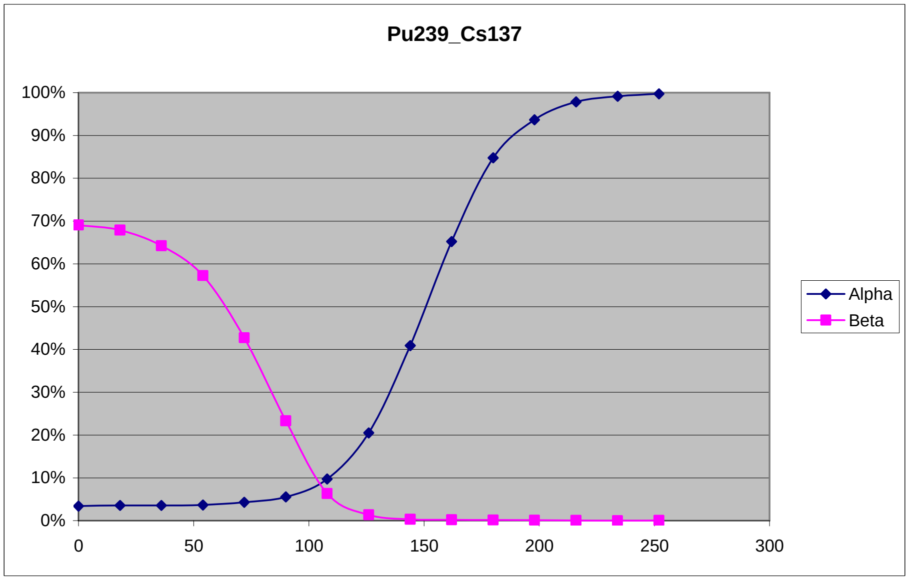
	% Spillover	
Setting	Alpha	Beta
0	2.9%	9.1%
18	2.6%	8.1%
36	3.2%	9.3%
54	3.2%	8.7%
72	3.6%	7.8%
90	4.7%	6.4%
108	7.3%	2.1%
126	14.0%	0.9%
144	34.5%	0.5%
162	62.2%	0.3%
180	84.1%	0.2%
198	94.2%	0.1%
216	98.2%	0.1%
234	99.4%	0.1%
252	99.8%	0.0%
Optimum:		
93	4.91%	5.21%



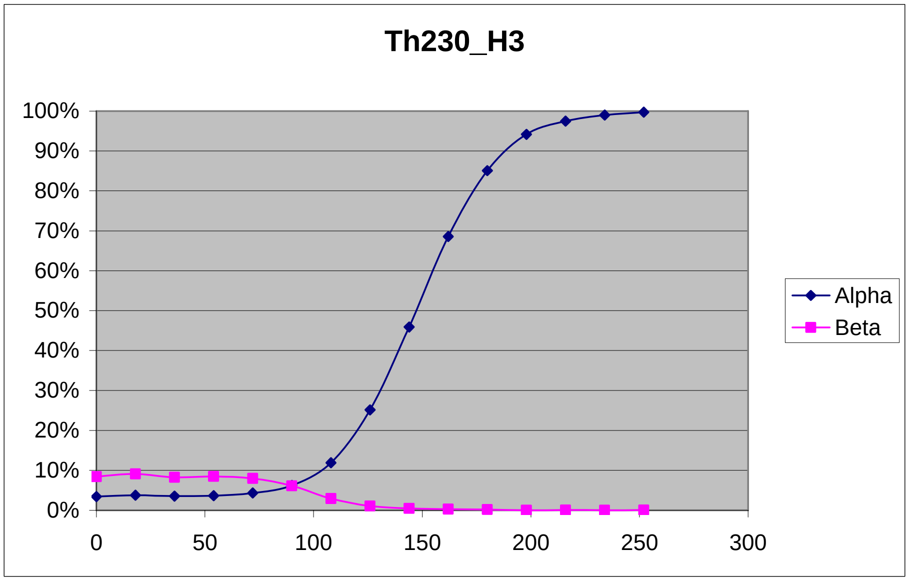
	% Spillover	
Setting	Alpha	Beta
0	3.4%	4.0%
18	3.6%	3.4%
36	3.9%	4.0%
54	4.0%	3.9%
72	4.7%	3.1%
90	6.4%	2.5%
108	11.7%	1.4%
126	24.7%	0.4%
144	46.3%	0.2%
162	69.1%	0.1%
180	84.7%	0.0%
198	93.7%	0.0%
216	97.3%	0.0%
234	99.1%	0.0%
252	99.5%	0.0%
Optimum:		
45	3.91%	4.02%



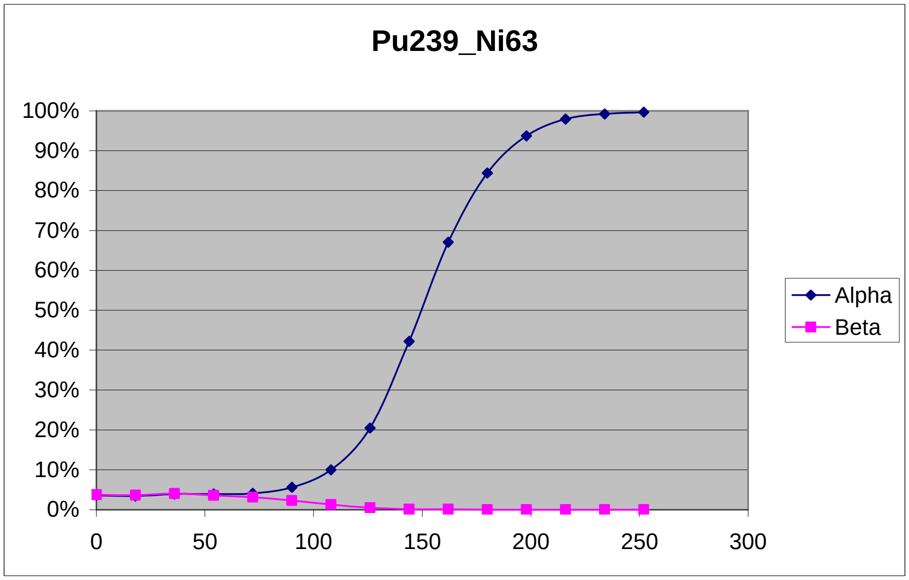
	% Spillover	
Setting	Alpha	Beta
0	3.4%	69.1%
18	3.6%	67.9%
36	3.6%	64.2%
54	3.7%	57.3%
72	4.3%	42.8%
90	5.6%	23.4%
108	9.7%	6.4%
126	20.5%	1.4%
144	40.9%	0.3%
162	65.2%	0.2%
180	84.8%	0.2%
198	93.7%	0.1%
216	97.8%	0.1%
234	99.2%	0.0%
252	99.8%	0.0%
Optimum:		
105	8.70%	8.68%



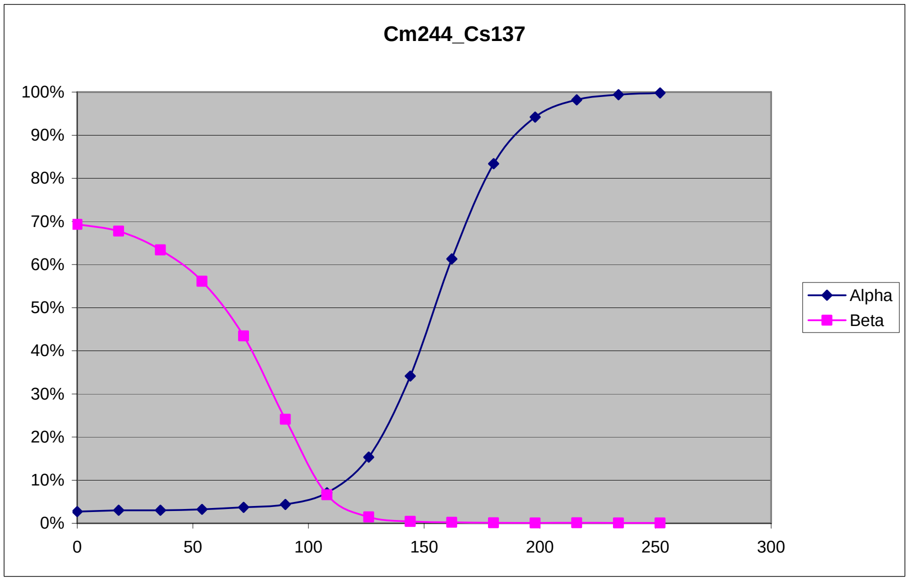
	% Spillover	
Setting	Alpha	Beta
0	3.4%	8.4%
18	3.8%	9.1%
36	3.5%	8.3%
54	3.7%	8.5%
72	4.3%	8.0%
90	6.2%	6.1%
108	11.9%	2.9%
126	25.1%	1.1%
144	45.9%	0.4%
162	68.6%	0.3%
180	85.1%	0.2%
198	94.2%	0.0%
216	97.5%	0.0%
234	99.0%	0.0%
252	99.7%	0.1%
Optimum:		
90	6.21%	6.14%



	% Spillover	
Setting	Alpha	Beta
0	3.6%	3.8%
18	3.5%	3.7%
36	3.9%	4.1%
54	3.9%	3.6%
72	4.1%	3.1%
90	5.6%	2.3%
108	10.0%	1.3%
126	20.5%	0.5%
144	42.2%	0.1%
162	67.0%	0.1%
180	84.4%	0.0%
198	93.7%	0.0%
216	97.9%	0.0%
234	99.2%	0.0%
252	99.7%	0.0%
Optimum:		
41	3.95%	3.98%



	% Spillover	
Setting	Alpha	Beta
0	2.7%	69.3%
18	3.0%	67.8%
36	3.0%	63.4%
54	3.2%	56.1%
72	3.7%	43.4%
90	4.4%	24.1%
108	7.1%	6.6%
126	15.3%	1.4%
144	34.1%	0.4%
162	61.3%	0.2%
180	83.4%	0.1%
198	94.2%	0.0%
216	98.2%	0.1%
234	99.4%	0.0%
252	99.8%	0.1%
Optimum:		
108	7.05%	6.59%



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