

CONFIRMING CRITICALITY SAFETY OF TRU WASTE WITH NEUTRON MEASUREMENTS AND RISK ANALYSES (U)

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ABSTRACT

The criticality safety of ^{239}Pu in 55-gallon drums stored in TRU waste containers (culverts) is confirmed using NDA neutron measurements and risk analyses. The neutron measurements yield a ^{239}Pu mass and k_{eff} for a culvert, which contains up to 14 drums. Conservative probabilistic risk analyses were developed for both drums and culverts. Overall ^{239}Pu mass estimates are less than a calculated safety limit of 2800 g per culvert. The largest measured k_{eff} is 0.904. The largest probability for a critical drum is 6.9×10^{-8} and that for a culvert is 1.72×10^{-7} . All examined suspect culverts, totaling 118 in number, are appraised as safe based on these observations.

INTRODUCTION

The Savannah River Site has stored TRU waste in 55-gallon drums for a number of years. Up to 14 drums are placed in 7 ft diameter by 7 ft tall concrete culverts. Before storage, the waste is assayed for fissile content to comply with conservative criticality limits. Assay techniques have evolved over the years. Initially only gamma measurements were used, but they later were supplemented by neutron coincidence measurements. Because higher ^{239}Pu concentrations in plutonium waste were estimated by these neutron measurements, it was important to confirm the safety of some of the earlier waste that was assayed only with gamma measurements.

The confirming studies required non-intrusive appraisals, including non-destructive assays (NDA) and risk analyses, as the culverts were subject to two severe administrative restrictions. The culverts could not be opened due to uncertainty about explosive potential from radiolytic hydrogen, and they were to remain stationary to exclude any potential criticality due to fissile material reconfiguration. The present study used NDA neutron measurements for estimating the ^{239}Pu and k_{eff} of a culvert and risk analyses for projecting its probability of ever becoming critical.

ANALYSES WITH NDA NEUTRON MEASUREMENTS

The NDA neutron measurements provided three analyses for appraising culvert criticality. The culvert ^{239}Pu masses were analyzed two ways: a direct method that essentially assumes mass proportionality with the neutron rates, and a statistical method that addresses neutron rate and mass fluctuations as a function of total mass. The measured neutron rates could also be examined in terms of subcritical multiplication to project k_{eff} for a culvert. The neutron measurements and the three analyses are described in detail in the following sections.

NEUTRON MEASUREMENTS

The NDA neutron measurements were used to examine suspect culverts and "calibration" culverts. A total of 118 suspect culverts were examined. Calibrations were performed with a test culvert into which a ^{239}Pu standard was placed at different drum locations. Additional calibrations used 36 culverts filled with waste that had been recently assayed with both the gamma and neutron coincidence methods.

In each NDA neutron measurement, two detectors centered atop the culverts monitored fast and slow neutron rates for a minimum of 200 sec. The detectors, 1-in diameter 8-in long ^3He -filled proportional counters, operated with standard counting electronics. The fast neutron detector was enclosed within a 1.25-in thick polyethylene annulus with an outer shield of 30-mil cadmium; the slow neutron detector had no moderator/shielding. Each measurement was corrected for background rates, which were generally small.

Calibration measurements with the test culvert provided interpretation for the NDA neutron rates measured for the suspect culverts. A canned source of $\text{PuF}_4/\text{PuO}_2$, typical of most SRS waste, with 253.8 g ^{239}Pu was used in this work. This source was placed in one of the drums loaded into the test culvert, and the corresponding neutron rates were measured. These culvert calibrations addressed drum location and moderator loading. Figure 1 illustrates the counting geometry. Using the fast/slow ratio to characterize the moderator, a calibration $C(x)$ in n/sec per gram ^{239}Pu results for each drum position x . Consequently, the measured neutron count rate r_n may be expressed as a sum of drum rates from the culvert,

$$r_n = M \sum C(x) m_n(x), \quad (1)$$

where M is the multiplication factor and $m_n(x)$ is the ^{239}Pu mass in the drum at x . The earlier gamma assay masses $m_r(x)$, which were suspected to be low, can be used to project r_p , an unmultiplied neutron rate,

$$r_p = \sum C(x) m_r(x). \quad (2)$$

The ratio r_n/r_p , as related to the above equations, form the basis of the culvert analyses for ^{239}Pu and k_{eff} .

The data reduction required scrutiny in arriving at r_n and r_p . Some of the measured rates had to be corrected for known ^{238}Pu contributions, and this was accomplished using test culvert calibrations with ^{238}Pu sources and measured correlations for the 36 calibration culverts. All suspect culverts were tightly stored on concrete pads, and thus backgrounds from adjacent culverts needed to be addressed. Measurements of backgrounds in a few empty culverts indicated that this background was relatively small, and it could be inferred by requiring agreement for the measured and projected fast/slow ratios. The $C(\mathbf{x})$ were deduced as the exponential average of two calibrations at the top and bottom of the drum, based on measured exponential behavior. Finally, drum location $\mathbf{x}$ information was limited to knowing whether a drum was in the top or bottom tier within the culvert. Fortunately, the neutron detectors (centered atop the culvert) view the hexagonal pattern of the seven drums symmetrically for each tier. Thus, the $C(\mathbf{x})$ of the six outer drums are identical and only that of the central drum differs - but not greatly. Calculations for r_p yielded an average of all possible loadings, along with a relatively small standard deviation.

DIRECT ANALYSIS FOR PU-239 MASS ESTIMATES

An estimate for the measured culvert mass $m_n = \sum m_n(\mathbf{x})$ is given by

$$m_n = (r_n/r_p) \sum m_r(\mathbf{x}) = (r_n/r_p) m_r, \quad (3)$$

which is deduced from Equations 1 and 2 by setting $M = 1$ and neglecting the $C(\mathbf{x})$ weighting factors in the sums. Because any real concern with criticality would imply $M \gg 1$, the above estimate is considered conservative. Furthermore, statistical fluctuations in r_n/r_p for culverts containing only ^{239}Pu were used to estimate the corresponding r_n/r_p upper limit for 0.1% excursions. The results in Table 1 all used such upper limits; however, most of the culverts required ^{238}Pu corrections in addition, for which the 0.1% upper limit could be modified. In any event, the combination of setting $M = 1$ and using this upper limit correction strongly argue that the estimates are conservative.

STATISTICAL ANALYSIS FOR PU-239 MASS ESTIMATES

Measurements for the culverts with the most recorded ^{239}Pu yielded r_n/r_p with the smallest fluctuations. This is consistent with the Central Limit Theorem, as the sum of many m_r measurements yields a smaller percentage error than an individual m_r measurement. Using r_n/r_p measurements of 27 culverts that contained only ^{239}Pu , a plot of $Y = \log(r_n/r_p)$ vs $X = \log(m_r)$ yielded an optimum chi-squared linear fit of

$$\sum_{i=1}^{27} (\delta Y_i / \sigma_i)^2 = N - 2 = 25 \quad (4)$$

when the distribution σ is proportional to $(82.3/m_r)^{-1/2}$. Using this σ to model a 0.1% upper limit for r_n/r_p yields the mass limits of the upper curve in Figure 2, which addresses the 118 culverts studied. These 0.1% upper limits correspond to r_n/r_p of 13 for $m_r = 82.3$ g, to comply with the factors observed between neutron coincident and gamma assays of waste samples or cuts¹.

A typical drum loading comprises 10-20 cuts, and thus the above experimentally deduced unit cut of 82.3 g of ²³⁹Pu is probably an overestimate. Indeed, typical cut data records for the earlier gamma assays indicate much smaller units. Such ²³⁹Pu gamma assays for the cuts of 68 drums were used to predict the corresponding neutron coincidence assay masses, using a scatter plot correlation between the two assays¹. These predicted neutron coincident assays for cuts were summed for their drum loadings and the corresponding error. A 0.1% upper limit for these drum results was propagated to predict a 0.1% upper limit for the culvert loadings, yielding the results of the lower curve of Figure 2.

The method using r_n/r_p fluctuations and the one using the transformed cut data were combined as a more realistic estimate, as illustrated in the intermediate curve of Figure 2. The former method is expected to be an overestimate because of the large unit mass for a cut. In the latter method, the cuts appear to favor smaller ²³⁹Pu masses; however, comparison with the assays of the 36 calibration culverts implied that the method was reasonable. In any event, small ²³⁹Pu drum loadings will have smaller cuts, so that estimates for low culvert loadings should be accurate. For the higher loadings, predictions of the r_n/r_p fluctuations should be used to assure conservative estimates. To match these conditions the transformed cut results were normalized to agree with the maxima of the r_n/r_p fluctuation results, as shown in Figure 2.

ANALYSIS BASED ON SUBCRITICAL MULTIPLICATION

The present study has implied that $m_n(x) > m_r(x)$. If this is true at all, Equations 1 and 2 predict that

$$r_n/r_p > M = 1/(1-k_{eff}) \quad (5)$$

Thus, r_n/r_p provides a conservative estimate of M , which leads to a conservative estimate of k_{eff} . The largest k_{eff} estimates for these culverts are reasonably low, as shown in Table 1. For culverts with $m_r < 500$ g, the minimum possible critical mass, r_n/r_p was multiplied by $m_r/500$ to yield the highest k_{eff} for a possible critical mass. These predictions are based on the same conservative r_n/r_p used for the direct analysis of ²³⁹Pu above; thus, the resulting k_{eff} are also conservative estimates.

PROBABILISTIC RISK ANALYSES

Risk analyses were conducted for both drums and culverts. A probabilistic risk analysis was required to appraise individual drums, as the neutron measurements could not distinguish one drum from another. Future unloading of drums from the culverts will have potential for redistributing the contents of any drum being moved, and because the ^{239}Pu of a drum is confined to a smaller volume, its criticality assessment will be somewhat different than that of a stationary culvert. At the same time, the probabilistic risk analysis for the drum can be extended to treat the culvert, yielding yet another appraisal of its safety.

PROBABILITY OF CRITICAL DRUM

The probability of a drum being critical is

$$P_0(C) = \int_L^U p(C|m) f(m) dm \quad (6)$$

where $p(C|m)$ is the probability of a criticality for ^{239}Pu mass m , $f(m)dm$ is the incremental probability for having mass m , and the integral limits range from $L = 500$ g (minimum critical mass) to $U = 5000$ g (estimated maximum possible mass). Models were developed for $p(C|m)$ and $f(m)$. Table 1 includes $P_0(C)$ results for the drums with largest m_r , calculated with the methods described below.

The $p(C|m)$ used PRA concepts²⁻³ to conservatively model the effects of fissile mass, fuel and moderator density⁴, geometrical configuration, and poisons. Figure 3 gives the $p(C|m)$ derived in the course of these studies³. This $p(C|m)$ is a 4th-order polynomial in m , which increases from 0 at $m = 500$ g to 10^{-3} at 5000 g. In Equation 6, this growth in $p(C|m)$ is countered by the rapid decrease of $f(m)$.

The above $P(C|m)$ was essentially modeled as the product of three basic probabilities², which were integrated over a range of critical densities for ^{239}Pu in water moderator³, viz.

$$p(C|m) = \int_{D_{\min}}^{D_{\max}} p_p p_k g(D) dD, \quad (7)$$

where $g(D)dD$ is the differential probability for critical ^{239}Pu density in the moderator, p_k is the geometrical configuration probability, and p_p is the probability associated with poisons. The range limits ($D_{\min}, D_{\max}$) were from critical data for reflected spheres of ^{239}Pu in water solution⁴. The probability factors are modeled similarly to those developed by Chay², but additional conservatism is added. In particular, $g(D)dD$ was increased to address the possibility of lumped fuel criticality. Also, p_k was increased to assure conservative estimates for fuel/moderator

overlap. Finally, p_p was increased to almost unity to diminish credit for the poisons. The overall conservatism relative to two values derived from the work by Chay² is exhibited in Figure 3. Here the $p(C|m)$ appear conservatively larger than Chay estimates for $m > 530$ g. For example, the $p(C|m)$ at $m = 800$ g is larger by a factor of 6. Because $p(C|m) \equiv 0$ at $m = 500$ g, the higher Chay estimate for this case probably used an m slightly above 500 g.

The $f(m)$ model uses neutron coincidence $m = m_{nc}$ that are inferred from the measured correlation²

$$\ln(m_{nc}) = 1.14 \ln(m_r) - 0.28 \pm (\sigma = 0.74) \quad (8)$$

to project m from a gamma assay m_r . A typical drum comprises 10 assayed cuts with different m_r , each predicting its own log-normally distributed m per Equation 8. Thus, $f(m)$ incorporated a conservative model including such components.

PROBABILITY OF CRITICAL CULVERT

The probability of a culvert being critical is

$$P_c(C) = \Sigma P_d(C) (1 + \alpha) \quad (9)$$

where the summation is over the culvert drums and α accounts for interactions between drums. Estimates of $\Sigma P_d(C)$ and α were developed to yield a conservative $P_c(C)$. These estimates were developed by examining a culvert that had m_r of 1221.55 g, which was the highest recorded value. Table 1 includes $P_c(C)$ estimates for culverts with the largest m_r .

A conservative estimate of $\Sigma P_d(C)$ can be shown to be given by $\Sigma P_d(C+) = N_d(\max) P_d(C)_{\max}$, where $N_d(\max) = m_{rc}/m_{rd}(\max)$ is the equivalent number of drums of maximum mass $m_{rd}(\max)$ for the culvert and $P_d(C)_{\max}$ is its corresponding $P_d(C)$. For the culvert with $m_r = 1221.55$ g, a $\Sigma P_d(C+)$ of 9.22×10^{-8} was calculated. The corresponding $\Sigma P_d(C)$ for the individual drums is 2.20×10^{-8} , which is lower by a factor of 4.2.

A value of α was developed by examining the $P_c(C)$ for the culvert with 1221.55 g ²³⁹Pu, and dividing it by the $\Sigma P_d(C)$ of its individual drums, to yield an α from Equation 8. This culvert had $P_c(C)$ of $P_c(C+) = 1.74 \times 10^{-7}$ when calculated using Equation 6 for a drum with the volume of the 14 culvert drums. For this calculation, the σ was reduced by a factor of 3 to reflect the larger number of cuts involved; a factor of $(14)^{1/2}$ or 3.74 could be argued, so the correction is conservative. The estimate $P_c(C+)$ is also conservative because Equation 8 involves unrealistically large m_r cuts, making the corresponding m_{nc} of the culvert too large. For this culvert, Equation 9 yields an α of 0.887 using the above $P_c(C+)$ and $\Sigma P_d(C+)$. From this we may develop a general expression for other culverts as

$$\alpha = \alpha_1 n_d \quad (10)$$

where α_1 is the α for a single interaction of two drums, and n_0 is the average number of such interactions per drum in the culvert. In the present case, with 14 drums, $n_0 = 31/14$ yielding a corresponding $\alpha_1 = 0.40$.

In this work, the $P_c(C)$ was conservatively estimated from Equation 9, using $\sum P_d(C) = \sum P_d(C+)$ and $\alpha = 0.40$ with n_0 calculated for $N_0(\max)$ drums. For the culvert with $m_r = 1221.55$ g this estimate is 4 times greater than that obtained by summing the individual $P_d(C)$ of its $N_0 = 14$ drums. Also, because this maximum culvert represents the maximum probability for interactions, the resulting α should be more conservative for the other culverts.

DISCUSSION OF RESULTS

This study assesses that the suspect culverts and drums pose no significant nuclear safety hazard. Table 1 presents the criticality estimates for cases with the largest culvert masses m_r , largest drum masses m_r , and largest k_{eff} estimates. Here, the highest probability for a drum criticality is $P_d(C) = 6.9 \times 10^{-8}$ and the highest for a culvert is $P_d(C) = 1.72 \times 10^{-7}$. The largest estimate for k_{eff} is 0.904. These probability and k_{eff} results are considered to be acceptably low, as they were deduced with very conservative assumptions. Furthermore, the results should be conservative relative to the more representative conditional probabilities of $P_d(C|k_{eff})$ and $P_c(C|k_{eff})$, which couple the favorable estimates of k_{eff} into the analyses.

For the NDA neutron measurements, the ^{239}Pu mass estimates based on the statistical analysis are all below 2800 g, a safe operating limit derived for these culverts⁵. However, five of the mass estimates using the direct analysis exceed this limit. Later culvert measurements with a HPGe gamma detector⁶ indicate that other neutron sources cause these high numbers. Yet, even without correcting for these sources, the table indicates the following maxima for these five culverts: $k_{eff} = 0.904$, $P_d(C) = 5.6 \times 10^{-8}$, $P_c(C) = 1.32 \times 10^{-7}$, and 1841 g ^{239}Pu with the statistical method. These maxima are all acceptably low.

The NDA neutron results are also conservative because they assumed that the ratio r_n/r_p of measured and projected neutron rates was either a correction for the culvert mass or the subcritical multiplication factor. In reality, it will be a combination of both. For example, if the combination comprises equal factors for mass correction and multiplication, the ratio becomes $(r_n/r_p)^{1/2}$ for both the direct mass estimate and its k_{eff} . Applying this example to the culvert with the highest direct mass estimate in Table 1 ($^{239}\text{Pu} = 5224$ g, $k_{eff} = 0.904$) yields results that are appreciably lower ($^{239}\text{Pu} = 1332$ g, $k_{eff} = 0.745$). Although the present study did not explore how the factors for mass correction and multiplication would be partitioned within r_n/r_p ,

the above example illustrates that both the direct mass estimates and k_{eff} of Table 1 are noticeably conservative relative to this refinement.

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Table 1. Criticality Results for Worst Cases

Case	Recorded Pu-239, g		Estimated Pu-239, g		Criticality Values		
	maximum drum m_r	culvert total m_r	culvert direct	total m_n statistical	k_{eff}	$P_D(C)$ 10^{-6}	$P_C(C)$ 10^{-6}
Max	-----> culverts <-----						
1	131.95	1221.55	2016	2488	0.394	0.010	0.156
2	123.6	1131.993	1779	2368	0.364	0.007	0.105
3	103.12	976.34	1707	2154	0.428	0.002	0.037
4	141.81	972.15	2442	2113	0.602	0.015	0.172
5	113.25	955.32	1732	2102	0.448	0.004	0.057
6	106.97	948.097	1495	2105	0.367	0.003	0.043
7	135.45	913.87	1377	2014	0.336	0.012	0.130
8	101.15	861.775	2025	1967	0.574	0.002	0.029
9	144.57	841.4	1082	1891	0.222	0.017	0.155
10	101.91	811.31	1614	1890	0.497	0.002	0.028
11	118.92	805.73	1745	1874	0.538	0.005	0.062
12	125.15	797.84	3101 ^a	1841	0.743	0.007	0.076
13	97.35	760.166	2599	1801	0.708	0.002	0.021
14	119.57	722.698	930	1705	0.223	0.006	0.054
15	134.59	694.59	1288	1670	0.461	0.011	0.091
16	144.82	688.95	3033 ^a	1657	0.773	0.017	0.125
17	130.66	685.25	934	1645	0.266	0.009	0.077
18	126.2	677.78	1714	1649	0.605	0.008	0.065
19	149.18	608.096	782	1472	0.222	0.020	0.122
20	125.39	554.018	854	1414	0.351	0.007	0.050
Max	--> drums <-----						
1	187.04	340.105	437	935	<0.100	0.069	0.147
2	183.29	267.41	560	782	0.107	0.062	0.098
3	183.26	241.97	311	742	<0.100	0.062	0.088
4	183.1	358.68	738	901	0.322	0.062	0.144
5	182.81	299.027	385	850	<0.100	0.061	0.113
6	181.85	323.805	553	857	<0.100	0.059	0.123
7	181.61	247.51	318	759	<0.100	0.059	0.086
8	181.54	346.83	739	963	0.323	0.059	0.133
9	180.21	350.357	451	901	<0.100	0.057	0.131
10	180.1	301.462	388	881	<0.100	0.056	0.107
Max	-----> k_{eff} <-----						
1	176.2	339.472	5224 ^a	961	0.904	0.050	0.114
2	179.71	355.056	3875 ^a	970	0.870	0.056	0.132
3	162.02	322.1	3168 ^a	878	0.842	0.032	0.076
4	50.67	342.056	2313	1048	0.783	<0.001	<0.001
5	174.7	333.29	2015	884	0.752	0.048	0.108
6	170.18	325.73	1788	879	0.720	0.042	0.094
7	74.28	343.477	1529	1078	0.673	<0.001	0.002
8	178.76	356.008	1444	931	0.654	0.054	0.129
9	167.09	230.51	1373	729	0.635	0.038	0.056
10	44.697	88.345	1276	474	0.608	<0.001	<0.001

a) Estimate exceeds calculated safety limit of 2800 g.

Figure 1. Culvert Counting Geometry

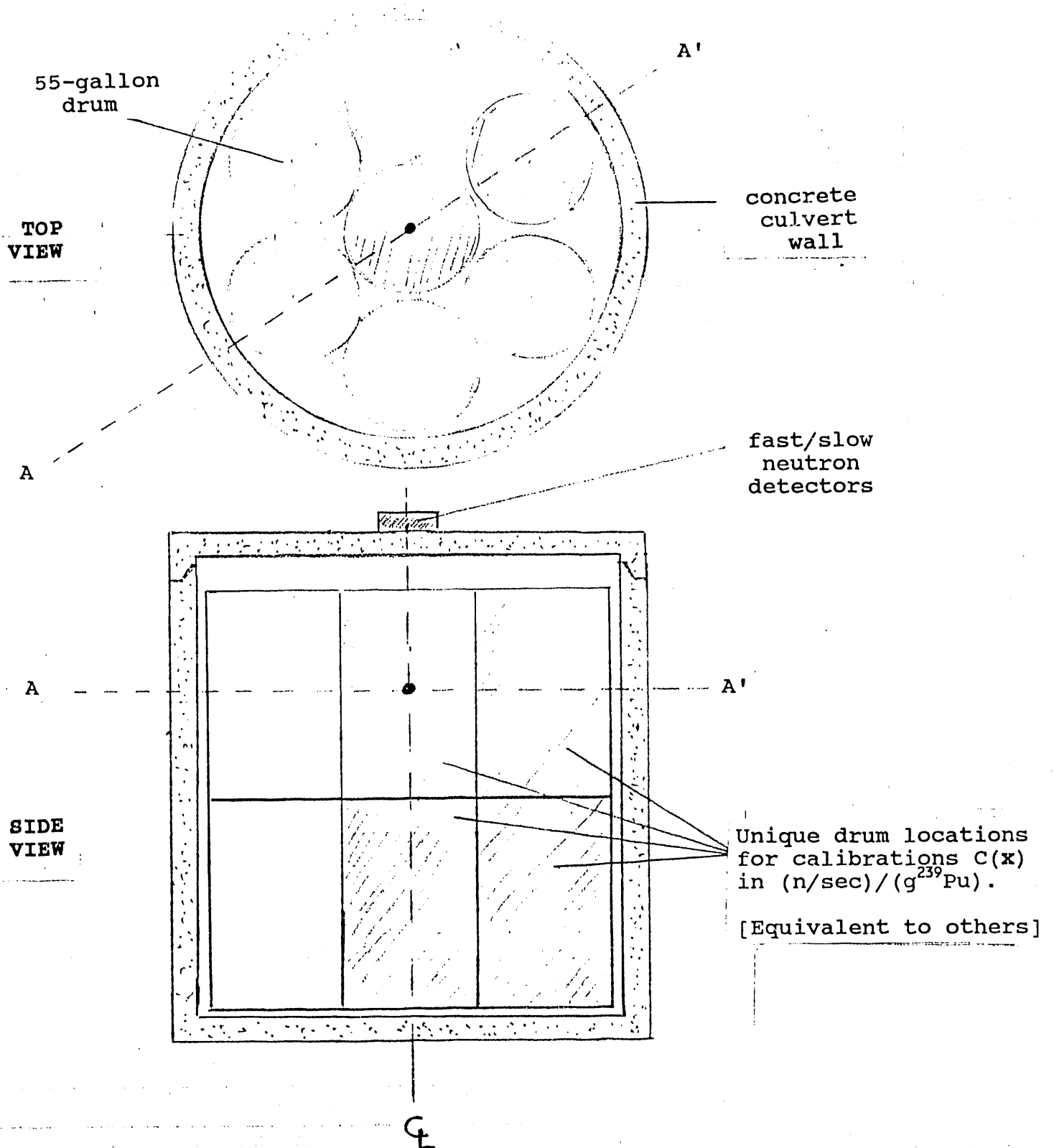


Figure 2. Statistical Estimates of ^{239}Pu in Culverts

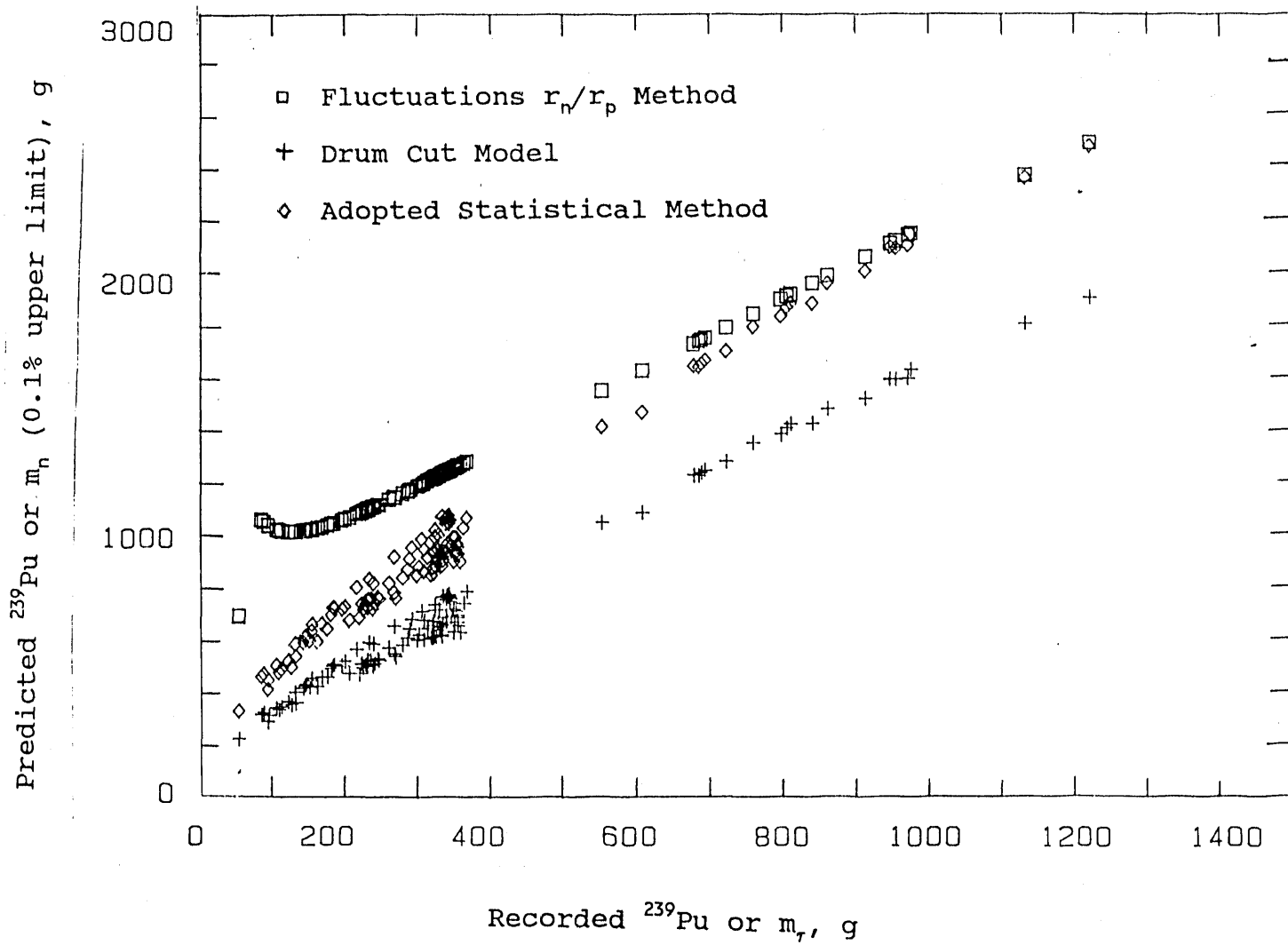
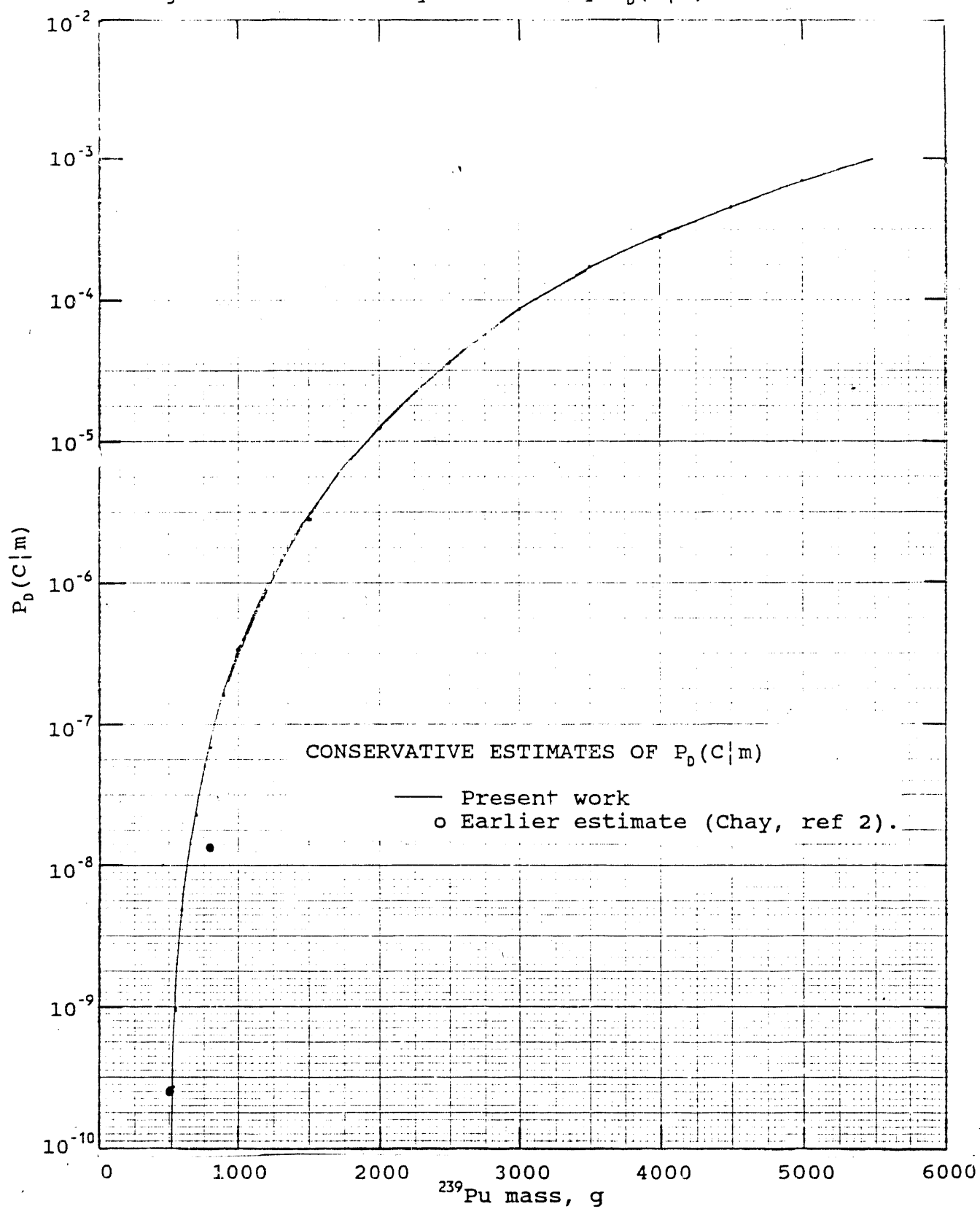


Figure 3. Criticality Probability $P_0(C|m)$ for Drums



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