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Title:

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RESULTS FROM LINC DETECTOR MCNP MODELLING

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SUMMARY

The response of the LINC detector to Cf and Pu sources has been determined using MCNP. The calculated results for Cf point sources agree well with measured values.
The 'base case' calibration (the response to Pu spread out over the likely volume of a crate containing a glovebox) is 2.24 Doubles/s/g ^{240}Pu. This, with the current background (1450 cps), gives a lower limit of detection of 5.3 g ^{239}Pu.
Several possibilities to improve the performance of the system have been investigated. Some looked initially promising, but measurements of the neutron background in the facility show that the neutron background is not only due to the stored drums in the same building. The neutrons arrive from all directions and so it would not be easy to shield the detector with a reasonable amount of neutron shielding. In addition the calculations show that thin cadmium sheets over the detector faces do not significantly reduce the background resulting from neutrons arriving from distant sources.
The performance of the detector for waste drums has been estimated. For such measurements it would be used at half its normal separation. This both increases the detection efficiency and reduces the background and results in a detection limit of 0.63g ^{239}Pu in weapons grade Pu (=114 nCi/g for a 1000lb drum) or 0.016g of pure ^{238}Pu = 0.28Ci.

1. INTRODUCTION

The purpose of this report is to describe some Monte Carlo calculations that were carried out to determine the performance of the LINC waste monitor (Ref. 1). Both MCNP and MCNPX were used (Ref. 2,3). Section 2 describes the model and section 3 describes some efficiency profile determinations that were made in order to compare with the detector as currently installed. Section 4 describes the expected response of the detector to plutonium and calculates the expected limit of detection. Section 5 deals with the investigation of methods to reduce the neutron background in order to improve the limit of detection. Section 6 describes the effect on the response from the changes introduced to reduce the background.

2. <u>MODEL</u>

^{3}He detectors
The ^{3}He detectors are 2 atmosphere, 2" diameter, and 72" active length, modeled with 0.5mm stainless steel walls.

Basic detector box
The basic detector box houses 5 ^{3}He tubes. The walls of the box and the cross-pieces which support the He tubes are made out of 0.5" thick polyethylene. The polyethylene density was taken to be 0.92 g/cm3. Two detector boxes are put together to form the detection panel for the device. The distance between the polyethylene faces of opposite panels of the device is 60". (= 56" between the covering panels (not modeled)).

Moderator
Four inches of polyethylene moderator surround the detector panels on all outside faces and the rear.

Floor
The bottom of the polyethylene moderator was 3" above the floor. The floor is covered with a 1" thick slab of polyethylene. Underneath this is a 0.25" steel plate on top of an asphalt floor. For the model the asphalt was replaced with a 30 cm thick sheet of concrete. The effect of this difference is expected to be small. (An early calculation showed that the difference in calculated efficiency between a model with concrete with a density of 2.5 g/cm3 and CaO_2 with a density of 1.0 g/cm3 was less than 0.5%.)
Cross sections of the model are shown in figures 1 and 2.

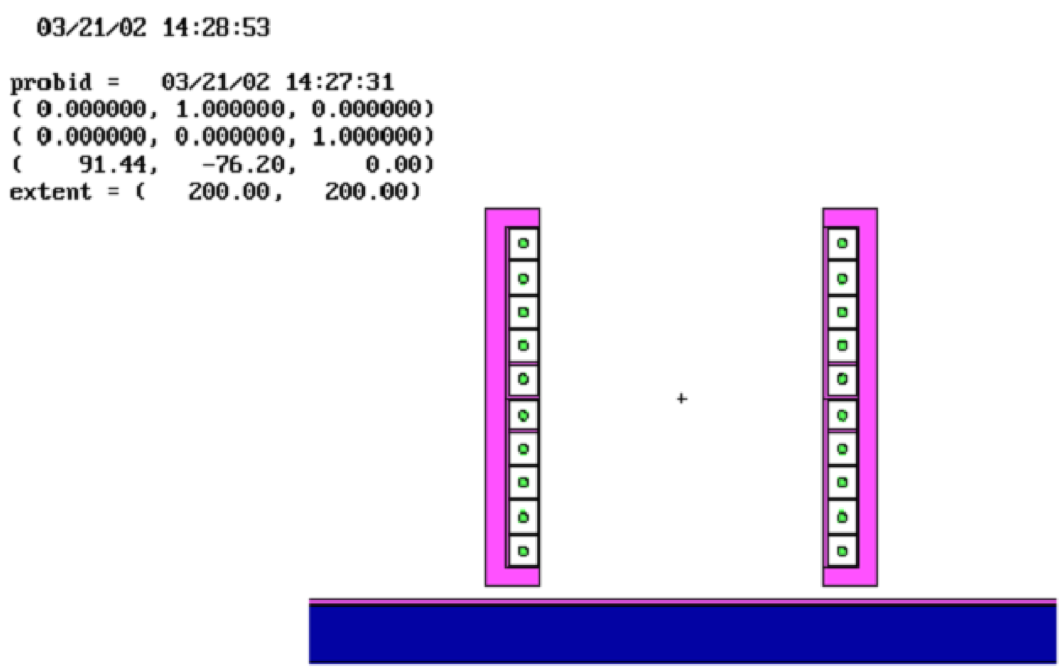

Figure 1 X cross section of model

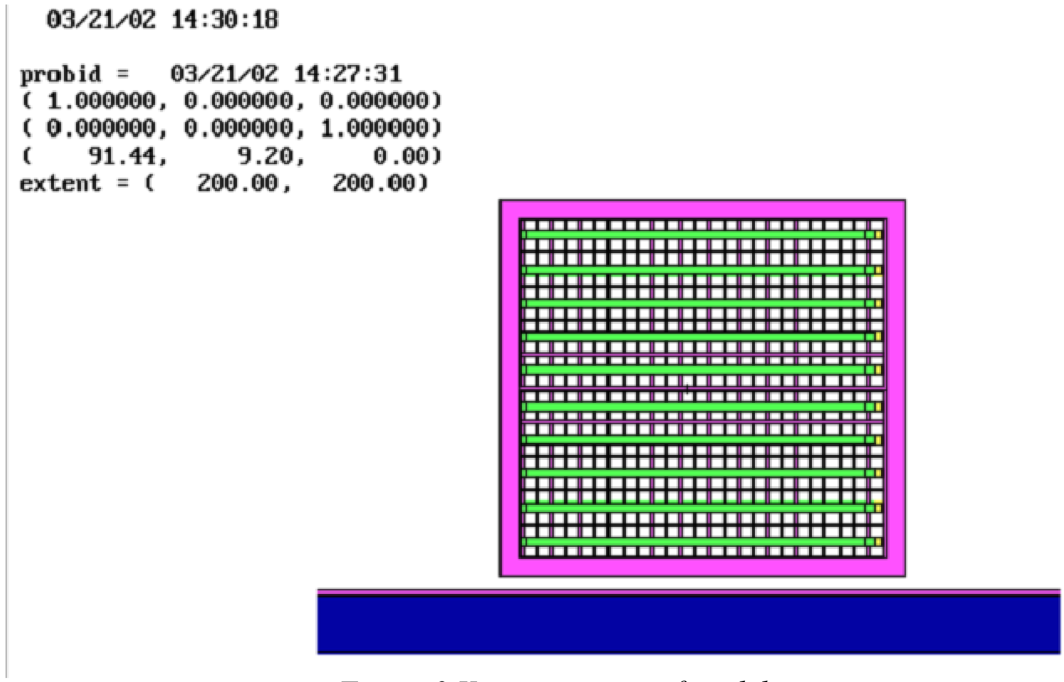

Figure 2 Y cross section of model

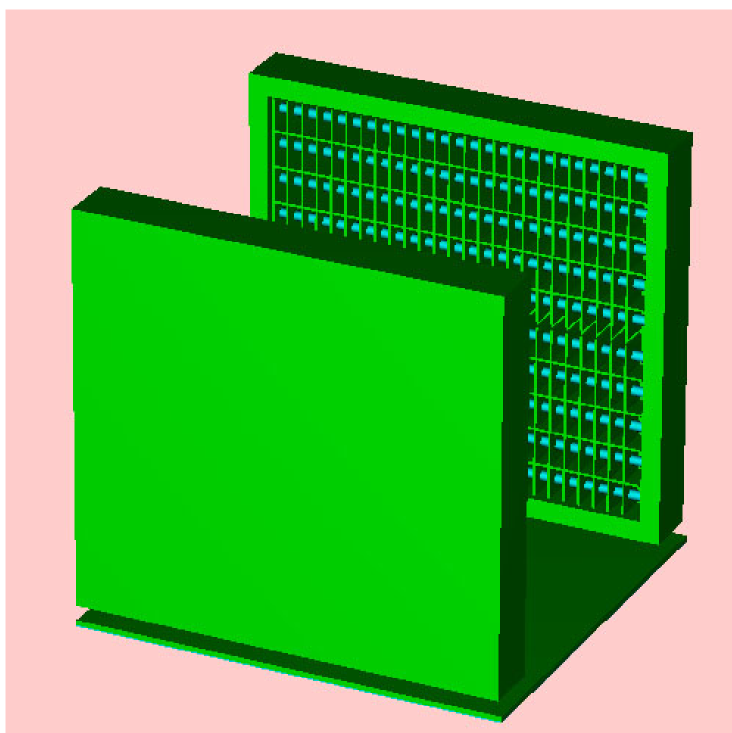

Figure 3. 3-D view of detector.

Figure 3 shows a 3-D view of the detector. The front polyethylene face of the far detector panel has been removed to show the polythene structure holding the ^{3}He tubes.

3. EFFICIENCY PROFILE RESULTS

The first set of calculations was done using a ^{252}Cf spectrum (average energy 2.3 MeV) to model the experimental measurements.

Central efficiency

The calculation of the Cf source located at the center of the chamber in the y and z directions and positioned in the center of the active region of the He tubes gave a total counting efficiency of 6.59%(.0026 fsd[1]). The measured efficiency with a central ^{252}Cf source was 6.7% (Ref. 1). The response as a function of time is plotted in figure 4. A least squares fit to the data gave a dieaway time of 240 μsec. This is caused primarily by the diffusion of thermal neutrons in the detector panels. The coincidence gate setting of the electronics is chosen to be somewhat larger than the dieaway time. The gate setting and the dieaway time determine the fraction of coincidence events that are detected.

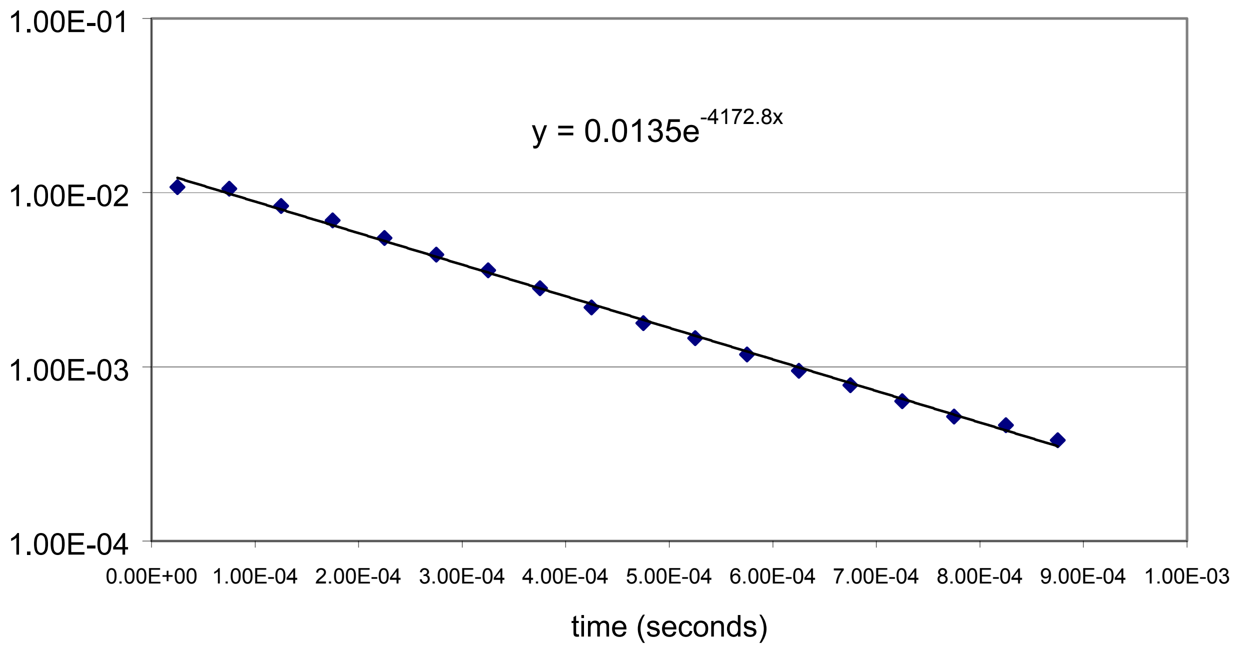

Figure 4. LINC dieaway = 240 usec

X direction profile

The profile in the x-direction was determined for a Cf point source in an empty detector. The results are given in table 1 and plotted in figure 5.

[1] fsd = fractional standard deviation of the calculated result

Table 1: X direction efficiency profile of LINC detector

Position (cm)	Total %	fsd
-50	2.28	0.0029
0	4.22	0.0026
50	5.99	0.0026
91.44	6.57	0.0051
150	5.72	0.0026
200	3.86	0.0027
250	2.05	0.0029

In each of the figures 5, 6 and 7, the dotted lines show the approximate extent of a 4' × 4' × 7' package located in the center of the cavity. (The uncertainty on each calculated point is usually smaller than the plotting symbol).

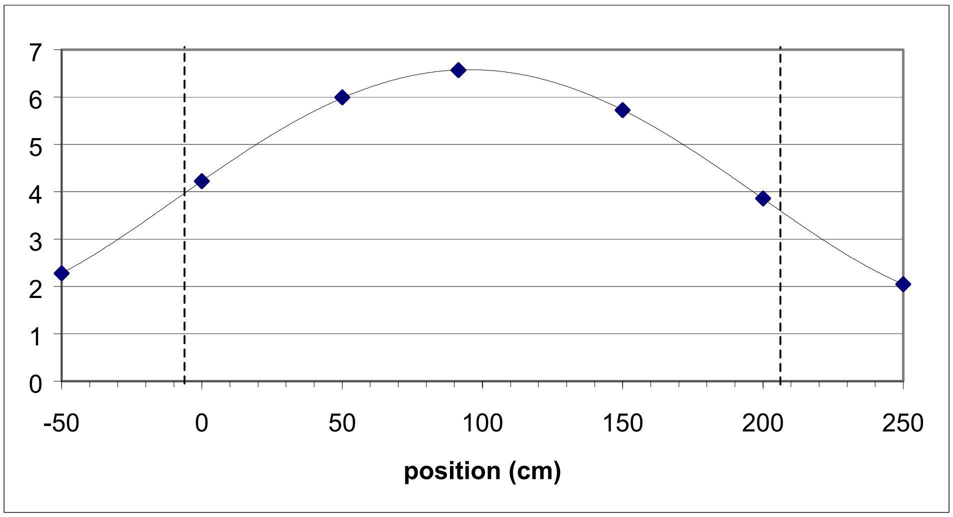

Figure 5. X efficiency profile

<u>Y direction profile</u>

The profile in the y-direction was determined for a Cf point source in an empty detector. The results are given in table 2 and plotted in figure 6.

Table 2: Y direction efficiency profile of LINC detector

position (cm)	Right Hand Panel %	Left Hand Panel %	Total %
-10	6.59	2.36	8.94
-20	5.98	2.37	8.35
-30	5.35	2.45	7.8
-50	4.29	2.7	6.98
-81.28	3.29	3.28	6.57
-100	2.74	4.18	6.92
-120	2.47	5.24	7.71
-140	2.35	6.47	8.83

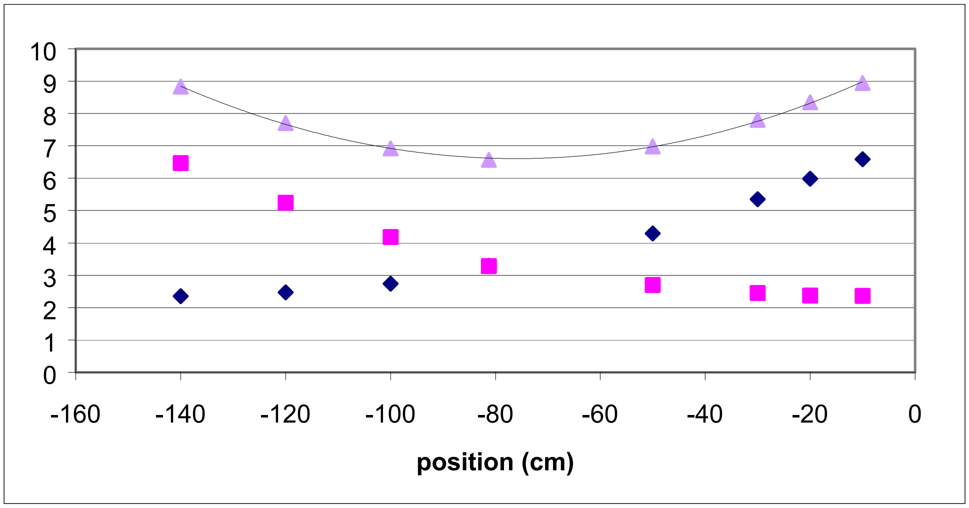

Figure 6. Y efficiency profile

The total and the separate contributions from the left hand and right hand panels are shown in the figure.

Z direction profile

The profile in the z-direction (vertically) was determined for a Cf point source in an empty detector. The results are given in table 3 and plotted in figure 7.

Table 3 Z direction efficiency profile of LINC detector

Position (cm)	Efficiency	fsd
-109	5.74	0.0051
-100	5.73	0.0052
-75	5.94	0.0052
-50	6.33	0.0052
0	6.57	0.0051
50	5.68	0.0051
75	4.83	0.0052
100	3.82	0.0052
125	2.75	0.0054
150	1.96	0.0056

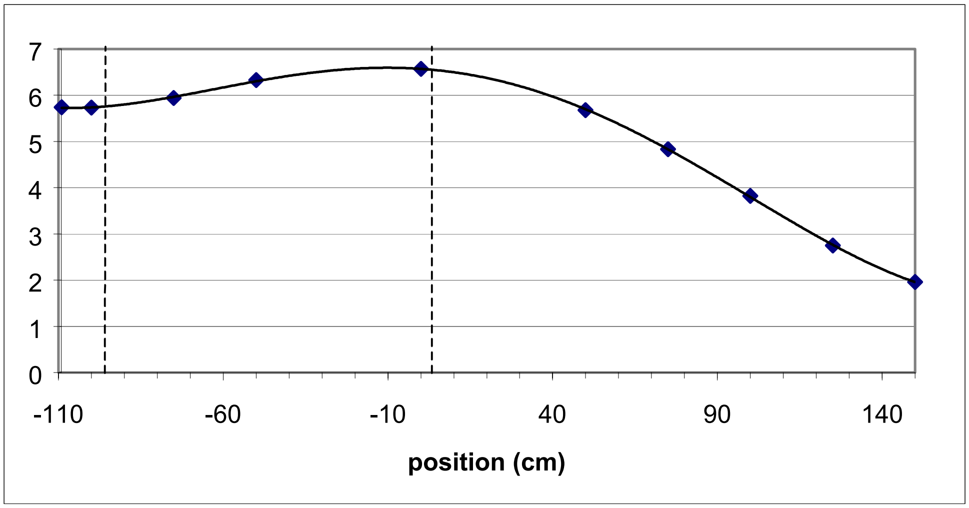

Figure 7. Z efficiency profile

4. CALCULATION OF DETECTOR PERFORMANCE I

The central efficiency was recalculated using a Watt fission spectrum for ^{240}Pu. In this case the mean energy is 1.98 MeV rather than 2.3 MeV as for ^{252}Cf. The total efficiency was 6.82% (.0024 fsd). This represents a (3.6 ± 0.4)% increase in efficiency for Pu relative to Cf in the center of an empty chamber.

Estimated Response

For a *non-multiplying* Pu sample at the center of the cavity, we can calculate the expected Doubles rate, D, from

$$D = mG\varepsilon^2 F\overline{\nu(\nu-1)}/2$$

where
m is the mass of $^{240}Pu_{eff}$,
G is the number of fissions/s/g ^{240}Pu
ε is the detection efficiency,
F is the fraction of events in the gate (=0.64 for a predelay of 4.5 µs and a gate of 256 µs with a dieaway of 240 µs), and
$\nu(\nu-1)$ is the 2nd factorial moment of the multiplicity distribution from ^{240}Pu.
This gives a coincidence rate of 2.68 Doubles/sec/g ^{240}Pu.

Estimated Performance

An important parameter of the system is the limit of detection (Ld), which depends on the uncertainty of the measurement. The background counting rate of the system can be determined with a particular uncertainty, σ_b. A measurement of an unknown amount of material is determined with an uncertainty σ_m (after background subtraction). The critical limit, Lc, of the system is a counting rate below which the container is said to be empty and above which the item is said to be not empty. Lc is determined by the acceptable false alarm rate. (A false alarm occurs when a measurement of an empty container, by chance, gives a value greater than Lc). Lc is obtained by multiplying σ_b by the appropriate factor for the desired false alarm rate. Commonly used factors for normally distributed measurements are shown in table 4. For example, for a 5% false alarm rate, Lc=1.645 σ_b.

Table 4. Factors for detection probability and false alarm rates for a normal distribution

False Alarm/Non-detection Probability	K, uncertainty multiplier for Normal distribution
0.05	1.645
0.01	2.327
0.005	2.575
0.001	3.090

The limit of detection of the system is greater than the critical limit by an amount that depends on the acceptable non-detection probability. For all measurements there is a certain probability that the result will, by chance, fall below Lc. As the mass of material increases this probability falls. Ld exceeds Lc by a factor times σ_m. The factors are the same as those for the false alarm probability and are shown in the above table. For example a non-detection probability of 5% is obtained with Ld = Lc + 1.645 σ_m.

Experimental Uncertainty

Experiments have shown for the present case $\sigma_b = 0.425 \pm 0.611$ for a single measurement of 1800 seconds and 0.425 ± 0.029 for the result of all 435 measurements. For measurements of very small quantities of material, the uncertainty of the measurement will be very close to that of the background, so that we take σ_m=0.612 (= sqrt($0.611^2 + 0.029^2$)) for an 1800 second measurement. Therefore for 5% false alarm probability and 5% non-detection probability we have Lc= 1.645×0.029 = 0.048 Doubles/sec and Ld = 0.048 + 1.645×0.612 = 1.054 Doubles/sec.

Using an estimated response of 2.68 Doubles/s/g ^{240}Pu we can convert Ld into units of ^{240}Pu: Ld=0.393 g ^{240}Pu. Assuming that the isotopic composition is 6% ^{240}Pu, this is equivalent to 6.55 g Pu or 6.16 g ^{239}Pu.

Increasing the counting time for a single measurement to 1 hour should reduce the uncertainty on the measurement to 0.433 ($\approx 0.612/\sqrt{2}$). This gives Ld=0.760 Doubles/sec, or 0.284 g^{240}Pu, 4.73 g Pu and 4.44g ^{239}Pu.

This limit of detection is almost completely determined by the singles counting rate of the device. If the singles background could be reduced to 50% of its present value, the limit of detection for a 1 hour measurement is expected to reduce to Ld = 0.048 + 1.645×0.216 = 0.403 Doubles/sec = 0.150 g^{240}Pu, 2.51 g Pu and 2.36 g ^{239}Pu.

Theoretical Uncertainty

The measured Doubles rate error is theoretically determined by the statistical properties of the coincidence counting method. The net doubles is determined from the difference of 2 quantities (R+A – A). Taking advantage of 'fast accidentals sampling', the theoretical value for the uncertainty can be calculated from $\sqrt{(\text{Accidentals/time})} = \sqrt{(\text{Singles}^2 \times \text{gate width/time})} = 0.547$ D/s. (using a singles rate of 1450 cps and an 1800s measurement). The measured error (given in the above section) is about 10% greater than this as other sources of variation such as any electronic variations and cosmic-ray fluctuations are included.

Table 5 shows the lower limit of detection, Ld, in terms of ^{239}Pu, for a response of 2.68 Doubles/g/^{240}Pu and a background totals level of 1450 cps (determined for a long measurement time) for a number of different sample measurement times.

Table 5. Lower Limit of Detection of ^{239}Pu as a function of measurement time.

Meas. time (s)	Mass ^{239}Pu (g) LLD
100	22.6
500	10.3
900	7.7
1800	5.5
3600	4.0

The calculations shown in the above table give the lower limit of detection, Ld, based on the theoretical distribution of the counting rate. As we have seen, these estimates are slightly more optimistic than the measured rates. A more complete estimate of the Ld can be obtained by making repeat counts of a surrogate sample that contains no plutonium. The observed standard deviation of many repeat measurements gives an experimental bases for the LLD.

In the above calculations, the effect of the counting rate from the item itself has been neglected. This will now be considered. If an item contains 8.4 g Pu (6% ^{240}Pu) then the neutron emission rate due to spontaneous fission of ^{240}Pu would be about 500 neutrons/sec. The detection efficiency is 6.82% and therefore the counting rate produced by the item is 35 cps. This can be neglected in the calculation of the measurement uncertainty.

If the material is in the form of oxide the neutron emission rate is increased by about a factor of 2 and the counting rate can still be neglected. If other light elements such as fluorine, boron or beryllium are present then the neutron production can be greatly increased. If the material was pure PuBe, the (α,n) rate would increased by a factor of over 1000 with respect to oxide and the totals counting rate would increase to greater than 25000 cps. This 'worst case' scenario would give an accidentals rate of 160000 cps and a corresponding lower limit of detection of 250 grams ^{239}Pu for a 1 hour measurement. Obviously this would preclude any categorization of drums at the required level.

5. INVESTIGATION OF BACKGROUND

The total counting rate of the detector when measured away from the facility was ~100 cps. In the facility this is increased to 1300-1400 cps, which has a big impact on the performance (see previous section). It is assumed that this increased rate is due to plutonium or other neutron source stored in the vicinity of the detector. In order to investigate this problem, the Monte Carlo model was used to calculate the detection probability for sources of neutrons stored in the neighborhood of the detector.

The neutron source was taken to be a point Pu source embedded in a sphere of polyethylene (at a low density of 0.4 g/cc) to represent the moderation of the material in which the Pu is contained. The source was located on the x-axis of the model at a distance of 16m (~50 ft) from the detector.
The first result gave a detection efficiency of 4.19×10^{-5} (.047). (If the moderating sphere is removed, the detection efficiency increases to 6.3×10^{-5}. This relatively small effect shows that the result is not too sensitive to the amount of moderator used in the model.) One observation is that in order to produce a counting rate of 1400 cps in the detector with this efficiency the neutron source strength would have to be 3.3×10^{7} neutron/s. If this was all Pu in the form of oxide, this would represent ~300 kg Pu. This suggests that other sources of neutrons may be in the area.

Several modifications to the system were considered in order to reduce the effect of these neutrons:

- Addition of 0.5mm cadmium skin in front of each the detector panels ("skin")
- Addition of 2" polyethylene sheet on the roof of the detector ("roof")
- Addition of a 2" polyethylene shield between the background source and the detector ("2" shield")
- Addition of a 4" polyethylene shield between the background source and the detector ("4" shield")

The results of these calculations are shown in table 6 and figure 8:

Table 6. Calculation of background rates for various shielding scenarios

	Efficiency x 10^{-5}	fsd	Estimated background
bare	4.19	0.047	1450
Cd skin	2.57	0.061	928
Roof + skin	2.62	0.065	944
2" shield (floor) + skin	1.74	0.058	661
4" shield + skin	1.15	0.090	471
4" shield (floor) + skin	1.10	0.094	454

In the case of the 4" shield, two entries are shown. The one marked "floor" indicates that the shield reached completely to the floor whereas in the other case the shield stopped at the same level as the bottom of the detector (3" above the floor). The difference is not large. The estimated background was calculated assuming that the count-rate was equal to 100 (cosmic background) + a contribution proportional to the calculated efficiency. This is then normalized to the current counting rate (1450 cps). The results are shown in the last column.

The Cd skins on the front of the detector panels reduce the background level to 64% of the initial rate. The addition of a 2” polyethylene roof (keeping the Cd skins in place) does not change the rate, which indicates that the contribution from scattering in the air above the detector is small.
The addition of a 2” shield (keeping the Cd skins in place) reduces the rate further to 46% of the initial rate. Increasing the thickness of the shield to 4” reduces the rate further to 33% of the initial rate.

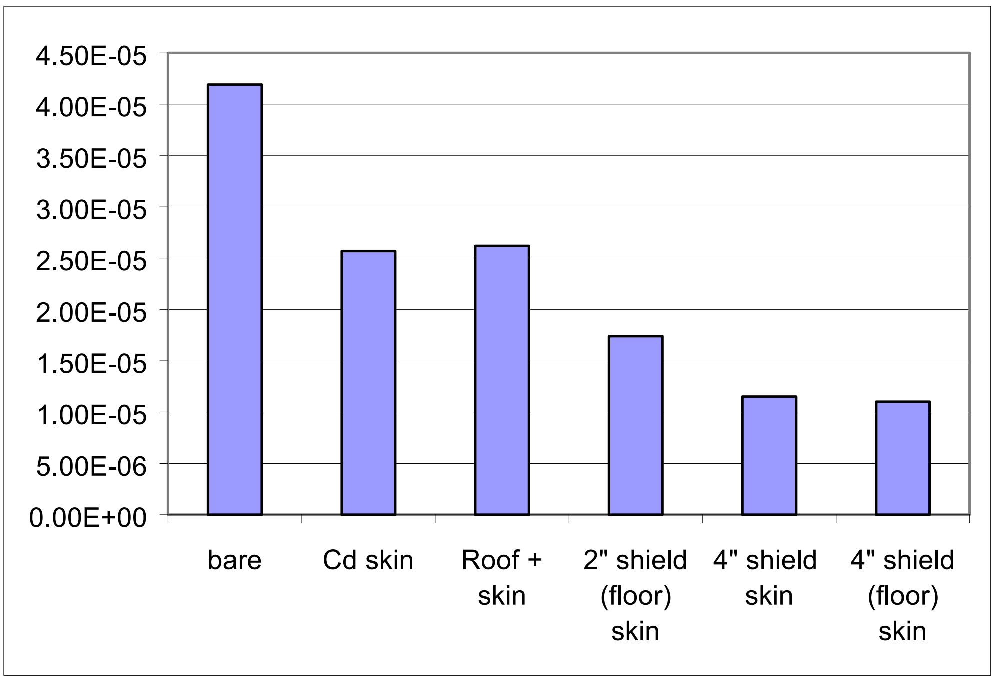

Figure 8: Background detection efficiency

The addition of Cd skins to the detector reduces the background but also changes the response to material inside the chamber. The detection efficiency and dieaway time are both affected. The response to Pu in the center of the cavity was recalculated for the Cd-covered case. The detection efficiency is 6.37% (.005) and the dieaway time (shown in figure 7) was 211 μs. These values change the response from 2.68 to 2.49 doubles/sec/g ^{240}Pu, a reduction of 7.5%. This change does not significantly modify the conclusions of section 4. For this calculation, the shield was not modeled. If it has an appreciable effect on the efficiency of a point source at the center of the detector cavity, it would be to increase the efficiency.

Table 7 shows the effect of the background reduction on the lower limit of detection (without taking into account the change in response from the Cd skins).

Table 7. Limit of Detection versus Singles Background Rate.

Background rate (Totals/s)	Uncertainty on net Doubles rate (D/s)	Lower limit of detection		
		^{240}Pu (g)	Pu (g)	^{239}Pu (g)
1450	0.39	0.26	4.3	4.0
928	0.25	0.17	2.8	2.7
661	0.18	0.13	2.1	2.0
471	0.13	0.09	1.6	1.5

These results for the additional shield will only be applicable to the real situation if the shield protects the detector from the complete source of the background. Subsequent measurements of the neutron background in the area showed that the neutron background was not principally due to neutron emission from material stored in the same building. This means that adding neutron shielding at one side of the detector will not be as effective in reducing the background as calculated above. An important question that remains is will the Cd skins be as effective at reducing the actual background as shown above. In order to answer this question more calculations were performed. The background model was changed to be a bare Pu source at a distance of 60 m (~200 ft) from the detector. Calculations were performed with and without Cd skins on the detector. The results are shown in table 8, together with the same case with a polyethylene sphere around the source, and a new calculation of the source in a polyethylene sphere at the original distance of 16m.

Table 8. Background Rate for source at 60m.

	Response No Cd	Response Cd	Ratio
60m bare	1.46e-5 (.036)	1.26e-5 (.040)	0.86
60m poly	5.90e-6 (.086)	4.62e-6 (.10)	0.78
16m poly	9.43e-5 (.026)	6.31e-5 (.032)	0.67

The results clearly show that the Cd skin is much less effective for the bare Pu source at 60m than for the moderated source at 16m. This indicates that there is a larger fraction of the signal coming from neutrons above the Cd cutoff. This may be due to air scattering. The implication of these results is that the scope for improving the performance of the detector is limited.

6. CALCULATION OF DETECTOR PERFORMANCE II

Base Case Pu calibration

The performance of the detector was calculated for a “base case” of a crate containing a glovebox. The crate is 4ft tall. There is an 8” false floor in the crate that will contain no material. The material was assumed to be uniformly spread vertically over 36” (leaving 4” empty zone at the top of the crate. (The vertical detection profile is fairly uniform and so the result will not be too sensitive to these assumptions). The crate is assumed to be centered along the length of the detector modules with Pu spread out over the central 6ft of the 7ft length. The crate is also assumed to be centered across the detector with material extending over the central 3ft out of the 4ft. The non-nuclear material in the glovebox is not modeled. This is a reasonable approximation for a relatively low density metal matrix. The total efficiency was calculated to be 6.22% (.0026). This is to be compared with a central point source efficiency of 6.82%, a relative reduction of about 9%. The calibration doubles rate becomes 2.24 D/s/g ^{240}Pu, giving a lower limit of detection of 5.26 g ^{239}Pu. This calibration is plotted in figure 9.

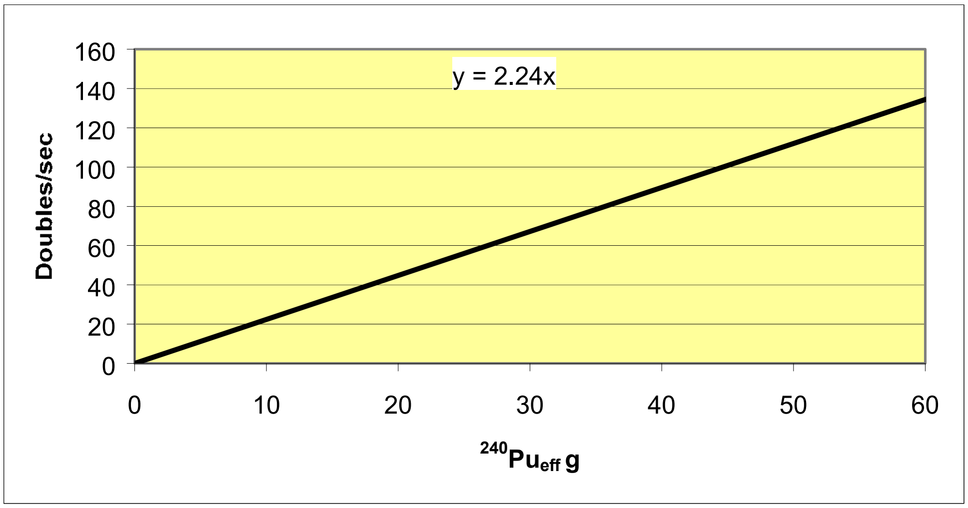

Figure 9. Base Case Doubles Calibration for LINC detector with Extended Source.

This is expected to be similar for crates of HEPA filters and other relatively low density matrices without too much moderation. Other specific cases can be calculated as necessary.

<u>Detection Limits and Isotopic Composition</u>

If we take the isotopic composition the same as used for Pu metal standards (Ref. 4.) we can calculate the mass of the Pu isotopes corresponding to an activity of 0.52 Ci. These are shown in table 9.

Table 9. Isotopic composition and Activity from Pu

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	Total
%	0.012	93.941	5.8523	0.1507	0.044	0.0551	
mass (g)	0.000826	6.467	0.40286	0.010374	0.003029	0.003793	6.88
Ci/g	17.12	0.06208	0.2271	0.002515	0.003929	3.422	
Ci	0.01414	0.4014	0.09149	0.00003	0.00001	0.01298	0.52

For this isotopic composition, 0.52 Ci corresponds to 6.5 g ^{239}Pu. (For 100% ^{239}Pu, 0.52 Ci corresponds to 8.37g).

We can also estimate the detection limit for pure ^{238}Pu. The coincidence response of ^{238}Pu is approximately 2.52 greater than ^{240}Pu. Thus the limit of detection with the system with its current background is 0.76/2.52/2.24= 0.135 g ^{238}Pu, which corresponds to 2.3 Ci. In order for the system to reach a limit of detection equal to 0.52Ci of ^{238}Pu it would be necessary to reduce the background to less than 280 cps (from 1450 currently).

Measurements of Other Containers

We can make a rough estimate of the performance of the device for other samples. For example to measure a 55 gal drum, the detector slabs could be moved to a separation of 30" (instead of 60"). The detection efficiency for a point Pu source becomes 12.5% and we could expect the background to drop to 50% of its current value. The detection limit then becomes 0.04g ^{240}Pu or 0.63g ^{239}Pu. This ignores the effect of the matrix (which could increase the detection efficiency if it was metallic). The actual background rate at this separation would need to be measured to confirm this performance. Assuming that the drum weighed 1000lbs, the detection limit corresponds to about 114 nCi/g.

For the case of a drum containing only ^{238}Pu then the detection limit is 0.016g ^{238}Pu, which has an activity of 0.28Ci.

If measurements of this type would be useful, it would be necessary to make a mechanical modification to the installation to allow the detector to be moved to this close separation.

7. REFERENCES

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