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HOLDUP MEASUREMENTS OF PLUTONIUM IN GLOVE BOX EXHAUSTS*

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ABSTRACT

A new measurement technique has been developed to quantify plutonium in process glove box exhausts. The technique implemented at Rocky Flats Plant utilizes a shielded, collimated 0.5"x0.5" bismuth germanate (BGO) gamma-ray detector. Pairs of measurements are made at one foot intervals along the duct. One measurement is made with the detector viewing the bottom of the duct with the detector crystal approximately 2 inches from the duct surface. The second measurement is made on the top of the exhaust pipe with the detector crystal 2 inches from the top of the duct. When the detector is placed in the bottom assay position, the area of the holdup material is assumed to extend beyond the detector field of view. The concentration of plutonium in g/cm² is obtained from this bottom measurement. The deposit width is determined from a model developed to relate the deposit width to the ratio of the count rates measured at the two positions, above and below the duct. Once a deposit width has been calculated, it is multiplied by the concentration determined from the bottom measurement to yield a mass-per-unit-length at the duct location. Total plutonium mass is then determined by multiplying the duct length by the average of the mass-per-unit-length assays performed along the duct. The applicability of the technique is presented in a comparison of field measurement data to analysis results on material removed from the ducts.

INTRODUCTION

Glove-box exhaust systems are used to evacuate the glove-box atmosphere and to isolate it from the work place. These systems carry the glove-box exhaust to plenums where High Efficiency Particulate Air (HEPA) filters are employed to remove airborne particulates.

Many glove-box operations, specifically dry processes such as burning and grinding, often cause small particles to be drawn into the exhaust system. These particles, including a fraction of the nuclear material being processed, then accumulate in areas of lesser or poor circulation within the closed exhaust systems. Measurement of this nuclear material accumulation (holdup) is essential when addressing criticality safety, employee health and safety, public risk and nuclear materials control and accountability programs in process areas.

Prior to the Summer of 1989, non-quantitative gamma surveys were accomplished primarily for the purpose of radiation protection at the Rocky Flats Plant (RFP). SCIENTECH Inc., under contract to the Department of Energy, performed a criticality safety assessment (CSA) at RFP from July through September of 1989. The CSA Team utilized non-destructive assay (NDA) equipment to perform surveys of selected glove-box exhaust systems to estimate plutonium holdup. In response to recommendations resulting from this CSA Team assessment, the Safeguards Measurements Group at RFP was chartered to develop and implement a measurement program to evaluate nuclear material holdup in glove-box exhaust systems.

Because the holdup material is contained in a structure of relatively uniform cross-section, the method of choice to assay the material in the ducts is nondestructive assay (NDA) using a far-field, line-source model. Safeguards Measurements personnel at Rocky Flats conferred with the Safeguards Technology Group N-1 at Los Alamos National Laboratory (LANL) who had developed a high resolution gamma-ray detection system specifically for holdup measurements [1]. The methodology and computer software for data acquisitions utilizing point, line, and area source models to quantify plutonium holdup was transferred to RFP. Rocky Flats then procured the appropriate instrumentation to assemble high resolution systems using this technology.

CHOSEN METHOD OF ASSAY

These systems, employing the proven technique of far-field, line-geometry measurements, worked well. However, utilization of this equipment and technique was restricted due to the physical layout of the glove-box exhaust systems. Ducts to be

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measured could not be isolated from surrounding systems using the far-field method because of the proximity of other plutonium bearing exhausts or glove boxes. Since many of the exhausts have little physical access due to other glove-boxes or utilities-bearing hardware, the detector had to be placed close to the ducts, thus violating the far-field, line source model. Since these limitations exist in a majority of the process areas, a smaller, more portable detector system had to be utilized. Concurrently, a model was developed for the analysis when the detector is used at "contact" with the surface of the duct.

Safeguards Measurements, in conjunction with the RFP Nuclear Instrumentation Development Group, built a bismuth germanate (BGO) detector system for use in the restrictive confines of the process areas. In the process of refining the measurement technique, several options were considered for the location of the second measurement position.

One labor intensive option is to take a series of measurements with the detector viewing the bottom of the duct at increasing distances. The count rate in the detector is constant for an area source as the distance from the source is increased, provided the detector field-of-view is filled by the source. The detector position at which the count rate in the detector began to decline would indicate that the field-of-view was no longer full. Thus, if the detector field-of-view and the distance of the detector from the bottom of the duct is known, the width of the deposit could be determined for that measurement. The extreme for this technique would be when the detector is one duct diameter below the bottom measurement position, at which point the field of view for the detector is the entire duct diameter. This same measurement position is obtainable by placing the detector on top of the duct. Thus, the top contact position was deemed most appropriate because this placement technique ensures consistent detector positioning at each measurement location, which is easily reproducible and independent of duct size.

Therefore, the RFP measurement technique is to position the detector for an upward view of the deposit two inches below the duct, and for a downward view two inches above it. The two inch space, from the detector crystal to the duct surface, was chosen to accommodate the collimator and a one inch space for a lead background shield. The lead shield is used to determine the background at both the top and bottom measurement positions. In conjunction with Group N-1 at Los Alamos National Laboratory, a mathematical model was developed to relate the count rates of the two measurements at each location along the duct to the material deposit width in the duct.[3] The validity of this model was tested at LANL and became the basis of the assay technique used for holdup measurements at Rocky Flats Plant.

DETECTOR SYSTEM

Since the measurements were to be performed on ducts which are relatively inaccessible because of their height from the floor or proximity to other glove-box utilities, a detector system was built which was easily transportable. The detector system incorporates a low resolution BGO crystal, 0.5"x0.5", mounted on a 0.5 inch diameter phototube. This detector is housed in a lead shield and collimator with an outside diameter of 2 inches. The collimator has a 0.5-inch diameter aperture 1-inch long which provides a field-of-view with a diameter of 1.5 inches for the detector position on the bottom surface of the duct. In the top position, the collimator provides a detector field-of-view, on the bottom of the duct, approximately equal to the diameter of the duct. See Figure 1.

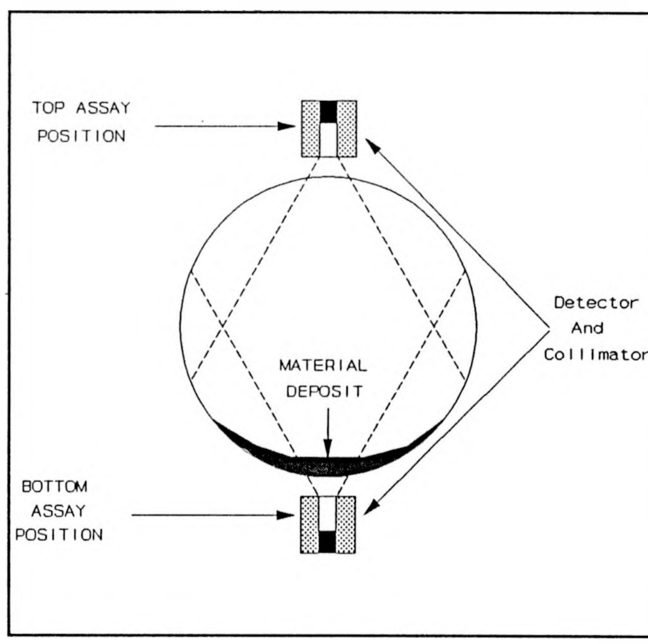

Figure 1: Duct cross-section showing position of detector for top and bottom measurements. Dashed lines indicate the field of view as defined by collimator.

A preamplifier circuit is built onto the detector base to give an overall detector and shield configuration which is light weight, provides shielding around the entire detector configuration and is housed in a package 2-inches in diameter by 8 inches long. Output of the preamplifier goes to a portable multichannel analyzer. The plutonium holdup is calculated from the intensity of the gamma-ray region of interest from approximately 300 to 450 keV. A second region of interest is established from 460 to 610 keV to correct for the

Compton continuum of high energy gamma rays in the plutonium analysis region of interest.

CALIBRATION

The BGO detector system is calibrated using a certified plutonium point-source standard. The point-source self absorption and encapsulation attenuation correction factors were determined by correlating the point source calibration data to data obtained measuring a set of well characterized line-source standards. The line-source standards consisted of plutonium oxide tightly packed in 18 inch long pieces of 3/16 inch aluminum tubing. The data for both the line sources and the point source were acquired using a high purity germanium detector. The 129, 203, 345, 375, and 414 keV gamma-ray energies from the decay of ^{239}Pu were analyzed. The correction factors for the point source were determined by comparing the attenuation corrected calibration for the line standards to the data for the point source. Since the 345, 375, and 414 keV gamma-rays fall within the region of interest assigned to the low resolution BGO system, a weighted average of these correction factors is applied. The point source is then used to generate an Area Calibration Constant (ACC) [2]. This constant is then used with the measurement data to calculate the concentration of Pu at the location measured.

DATA ACQUISITION

Data is obtained by positioning the detector to acquire pairs of measurements at specific intervals along the duct. Typical spacing between locations is 1 foot for ducts less than 14 inches in diameter and 2 feet for ducts greater than 14 inches in diameter. Normal data acquisition time for each of the four measurements (top background, top assay, bottom background, bottom assay) per location is 100 seconds live time. The data acquisition process is carried out using an integrated software package on a portable, personal computer interfaced to the multichannel analyzer that directs the operator through the measurement process.

DEPOSIT MODEL

The following assumptions are used in the analysis. For horizontal ducts, the majority of holdup material rests along the bottom of the duct. This assumption has been verified by field measurements and by a remote video camera used inside ducts targeted for clean-out. When the detector is placed in the bottom assay position, the area of the holdup material is assumed to extend beyond the detector field of view. This measurement is used to determine the concentration of the plutonium in gm/cm^2 at the measurement location. When the detector is placed in the top assay position, the holdup material may not fill the detector field of view but is assumed to be a uniform strip parallel to duct axis and centered on the detector field of view as illustrated in Figure 2.

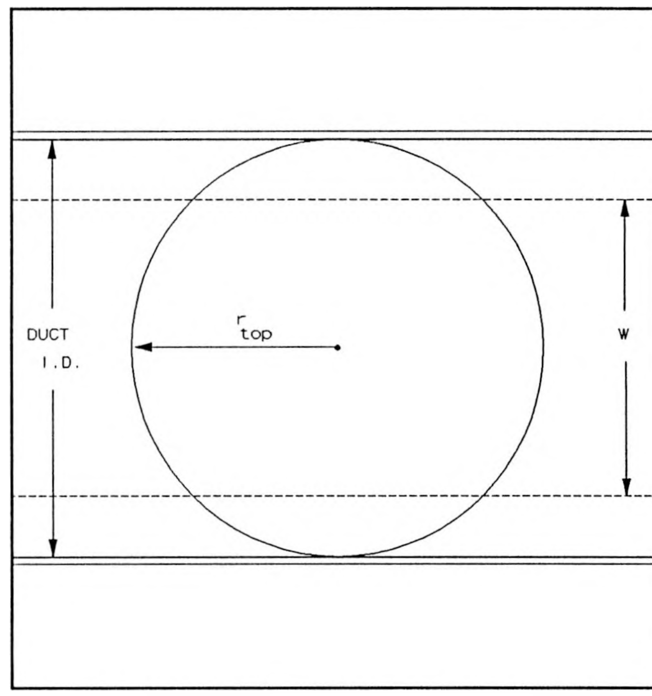

Figure 2: Field of view seen by detector in top position. W is the width of material on the bottom of duct; r_{top} is the radius of the field of view defined by collimator.

By using a ratio of the top count rate (C_T) to the bottom count rate (C_B), the width of the holdup material may be estimated using a mathematical model developed jointly by RFP and LANL personnel [3]. The width of the holdup material is defined as the extent (i.e. arc length) of material extending from side to side along the bottom of the duct transverse to the duct axis, i.e., W in Figure 2. The relationship between the count-rate ratio and the width is given by

$$\frac{C_T}{C_B} = \frac{2}{\Pi}\left[\sin^{-1}(X) + 2X\sqrt{1-X^3} + X^3 \ln\left(\frac{1-\sqrt{1-X^2}}{X}\right)\right] \quad (1)$$

where:

$X = W/2r_{top}$,

$2r_{top}$ = Detector field-of-view projected on duct surface.

The validity of this model was tested at Los Alamos National Laboratory by Group N-1 using thin ^{235}U foils nominally 46 cm long and either 3.8 or 7.6 cm wide. These foils were laid in the bottom of the pipes

NCR = Net Count Rate in counts per second for the bottom detector,

ACC = Area calibration constant in grams-seconds / cm^2,

W = Width of holdup material in centimeters,

CF_{Pipe} = Attenuation correction factor for the intervening material.

The point assay calculation is repeated for each measurement location along a duct. Each location can be described in terms of its relative position from the starting point. The plutonium mass (in grams) in the duct is given by

$$Pu\ (gms) = \sum_{i=1}^{n-1} \frac{(y_{i+1} + y_i)}{2} \times (x_{i+1} - x_i) \qquad (3)$$

Where:
n = number of assay locations,

y = point assay (gram/cm),

x = position of each point assay (cm) along the length of the duct.

DATA COMPARISON

Typically NDA measurements are validated by measuring standards representative of process material. This validation method is not easily accomplished for duct holdup measurements due to the number and variety of matrices encountered in holdup material. As an alternative, in areas where ducts are cleaned out, a comparison of before-clean-out and after-clean-out measurements can be made to proven NDA methods (calorimetric assay) or destructive analysis of the material removed.

RFP personnel have cleaned several sections of duct and the removed material has been assayed by calorimetry/gamma-spectroscopy techniques to ascertain the nuclear material content. These comparisons are available for four separate duct sections. Table 1 summarizes the delta measurement data, from before-clean-out data and after-clean-out data, to the corresponding measured values for the bulk material removed. These data show good agreement between the delta measurements and the analyses of the removed material. No difference greater than 21% is observed. In the measurement of Line 2, the before-clean-out measurements were biased high due to plutonium material stored adjacent to the measurement locations. This material was removed prior to the clean out operations and resulted in a higher delta measurement for the comparison results.

In an attempt to compare the contact-measurement technique to the far-field, line-source model, a 154 foot section of duct was measured with both the BGO detector system and a high resolution germanium detector system. The total holdup measured was 162 grams of plutonium for the BGO system and 189 grams for the line-source model using the germanium detector. The two methods showed good agreement except for two 5-foot sections which had higher amounts of plutonium in the line-source method where the exhausts of two vacuum pumps contaminated with plutonium could not be excluded from the field of view. Nevertheless, the results from the two measurement campaigns, using two different measurement methods and detector systems, differ by only 15%.

SUMMARY

These comparisons have exceeded expectations. The limited data available provide an indication that the BGO detector top/bottom contact measurement technique is valid. As additional data are accumulated, the overall effectiveness of the applicability of the top/bottom ratio model will be assessed.

Contact holdup measurements on glove-box exhaust ducts, employing the top/bottom width model is a viable method. It is especially useful in areas where pipe configuration is not conducive to the far-field line approximation method. It is also especially applicable in those areas where small amounts of nuclear material holdup make far-field, line-geometry measurements impossible due to the lack of gamma-ray activity emanating from the duct.

REFERENCES

1. P. A. Russo, R. Siebelist, J. A. Painter, and J. E. Gilmer, "Evaluation of High-Resolution Gamma-Ray Methods for Determination of Solid Plutonium Holdup in High-Throughput Bulk-Processing Equipment," Los Alamos National Laboratory report LA-11729-MS (January 1990).

2. N. Ensslin and H. A. Smith, Jr., "Attributes and Semi-quantitative Measurements, "Chapter 20 in Passive Non-destructive Assay of Nuclear Materials, NUREG/CR-5550, T. D. Reilly, N. Ensslin, and H. A. Smith, Jr., Eds., Los Alamos National Laboratory document LA-UR-90-732 (1991).

3. G. A. Sheppard, P. A. Russo, T. R.

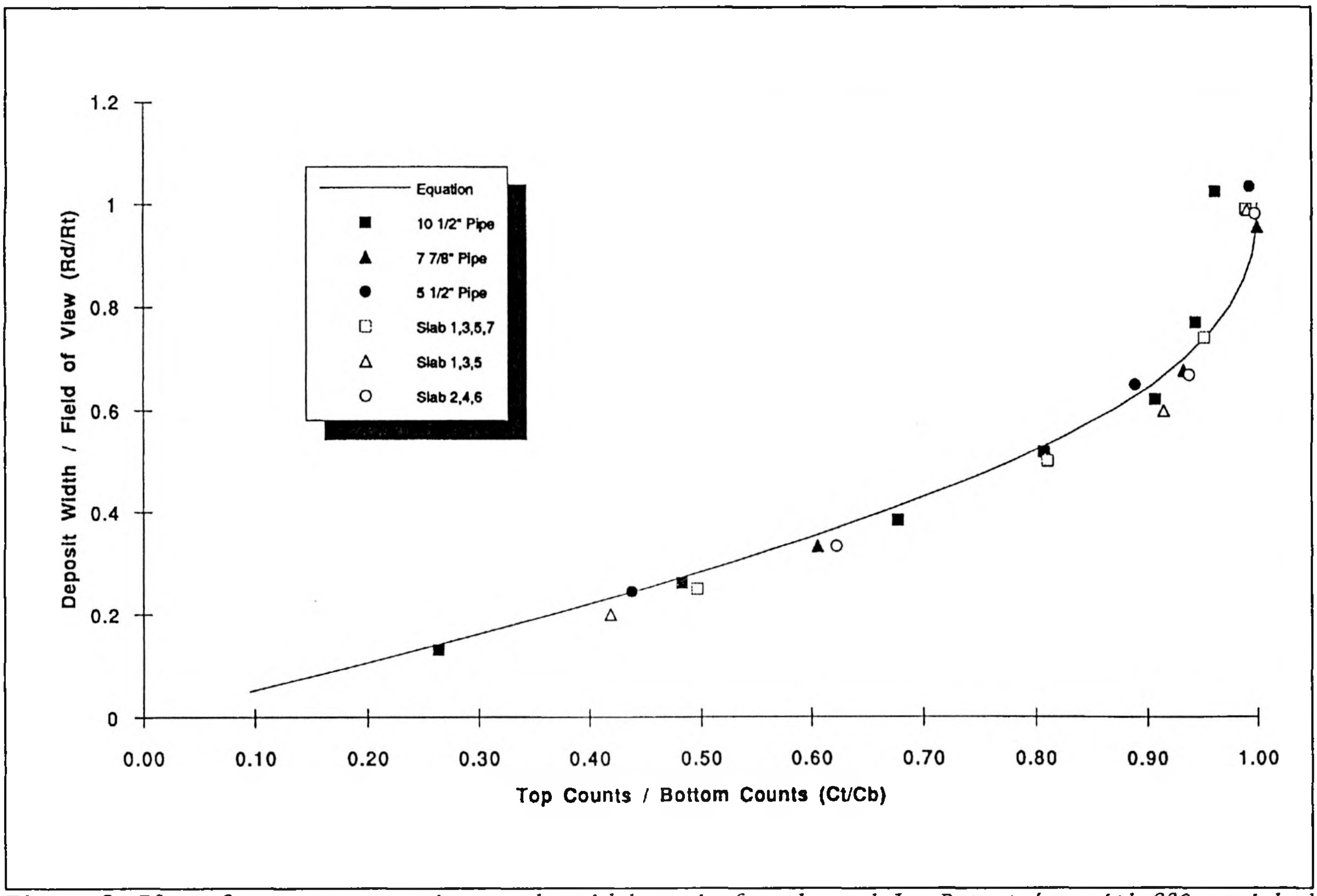

Figure 3: Plot of count rate ratio vs. the width ratio for the model. Data taken with 93%-enriched uranium foils arrayed in pipes of different diameters and on a flat surface (slab). Reference 3.

parallel to their axes. The intensity of the 186 keV gamma rays from uranium was measured with a sodium iodide detector using collimation that restricted the detector field of view to that of the BGO detector used at Rocky Flats. Results are shown in Figure 3 for three different simulated pipe diameters using as many different foil combinations as could be accommodated in each pipe to fill the detector field-of-view. In addition, data were taken with foils held in a plane, which more nearly describes the physical situation described by the model. The top-to bottom count rate ratios were computed for each measurement pair. The ratios of the actual deposit width and corresponding geometric field-of-view are plotted as a function of count rate ratio in the Figure 3. The solid symbols represent the ratios of the count rates for the foils in the pipe configuration while the open symbols represent the ratios of the count rates with the foils in the plane (slab) configuration. The solid line is the result computed from Equation 1. The width ratio predicted using the equation typically errs from the actual ratio of deposit width to field-of-view radius by less than 10%. It is important to note the relative insensitivity to deposit curvature. The model reliably predicts the deposit width regardless of pipe diameter or the curvature of the deposit.

Equation 1 is applicable for cases where C_B is greater than C_T. In cases where the ratio of the top to bottom count rates is not statistically different from 1, the holdup is assumed to be uniformly deposited around the inner surface of the duct, and the inner circumference of the duct is substituted for the width (W). This case is typically encountered in ducts with low levels of activity and in ducts from which the material has been vacuumed. This is an inherent difficulty with the clean out process since no attempt is made to remove material from the top interior of the duct. In the case of a vertical duct, the inner circumference of the duct is also substituted for the width (W).

Once the data have been collected for a duct section, a final holdup value is calculated by the data-acquisition software. For each measurement location, a point assay (PA) in grams per unit length of duct is calculated based on the net count rates, area calibration constant, material width, and a correction factor for attenuation by the duct wall.

$$PA\ (Gm/Cm) = NCR \times ACC \times W \times CF_{Pipe} \quad (2)$$

where:

Wenz, M. C. Miller, E. C. Piquette, F. X. Haas, J. B. Glick, and A. G. Garrett "Models For Gamma-Ray Holdup Measurements at Duct Contact," to be presented at the Institute of Nuclear Materials Management 32nd Annual Meeting; New Orleans, LA, July 28-31, 1991.

Table 1. Duct Clean-out Data

Duct	Duct Length (Ft)	Number of Points Measured	Delta Measurement (Grams Plutonium)	Measured Value (Grams Plutonium)
1	94	125	307	302
2	92	103	150	124
3	33	56	76	76
4	44	86	124	122

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Introduction

- How it All Started

  - Criticality Safety Assessment

  - Ducts Identified as a Potential Concern

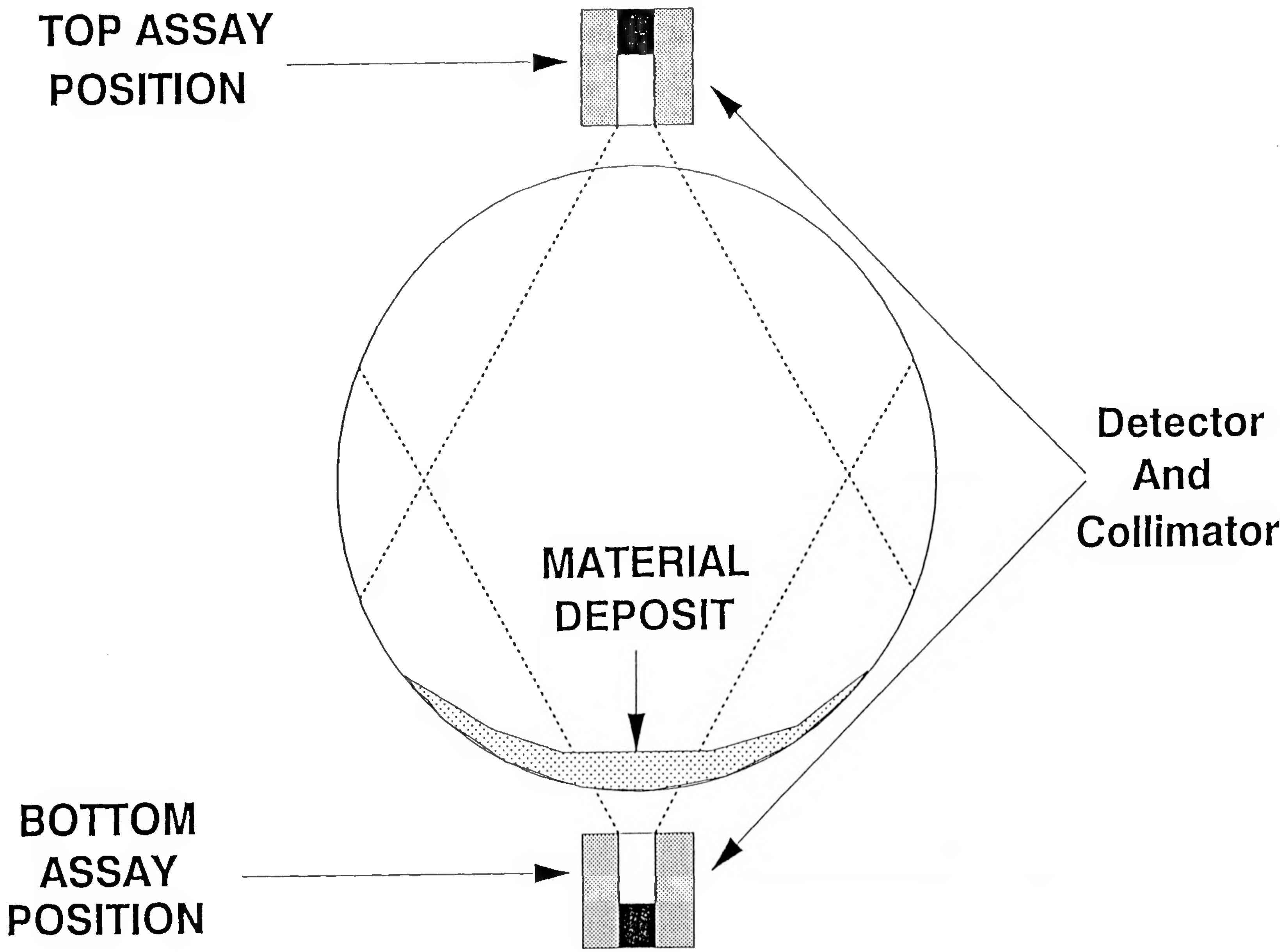
  - Chartered to Develop and Implement a Program

Detector System

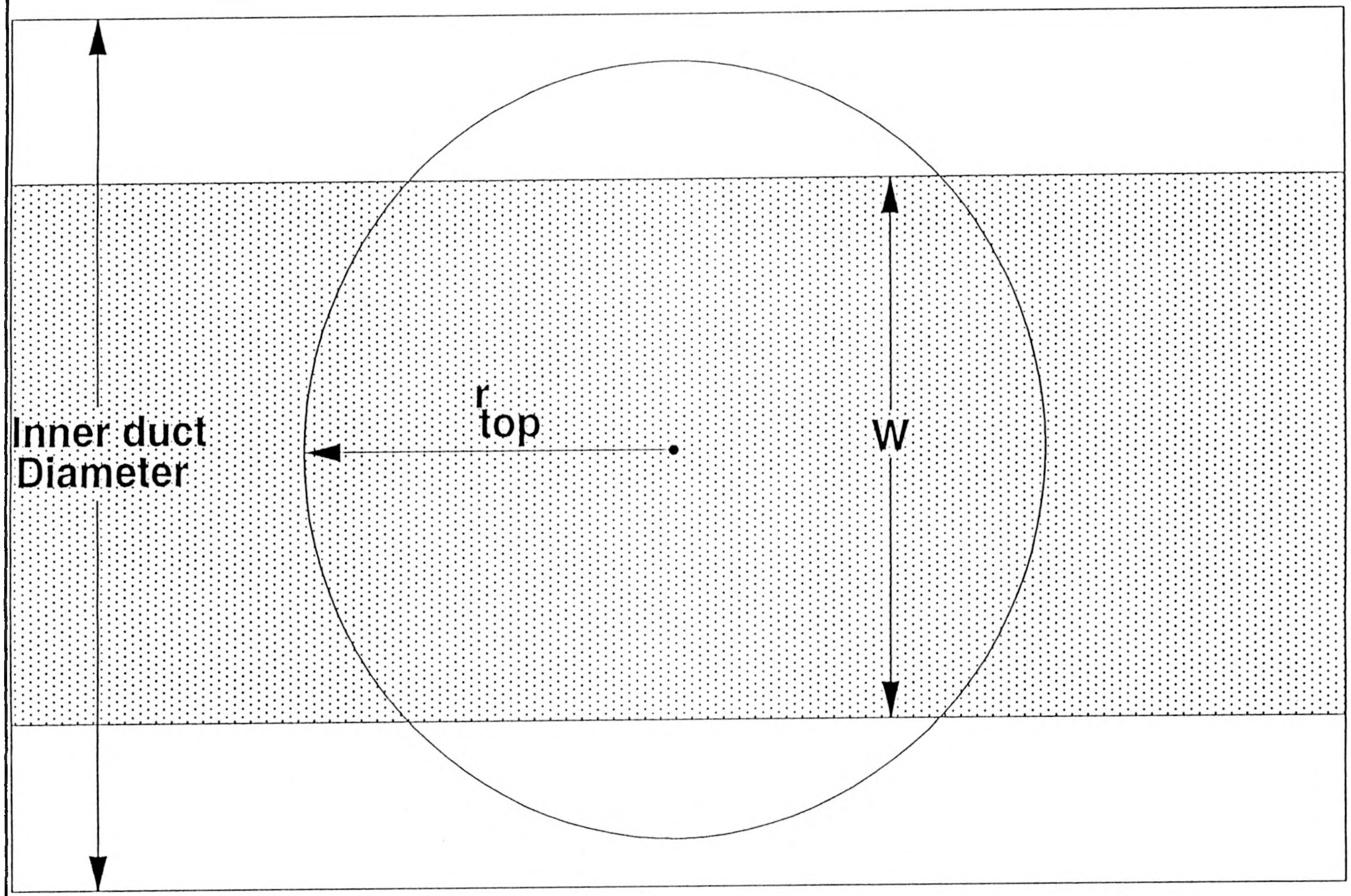
- Difficulties with Duct Configuration
- Contact Measurements Feasible
- Bismuth Germanate (BGO) Detectors Selected
- Collimated, Shielded System Easily Portable

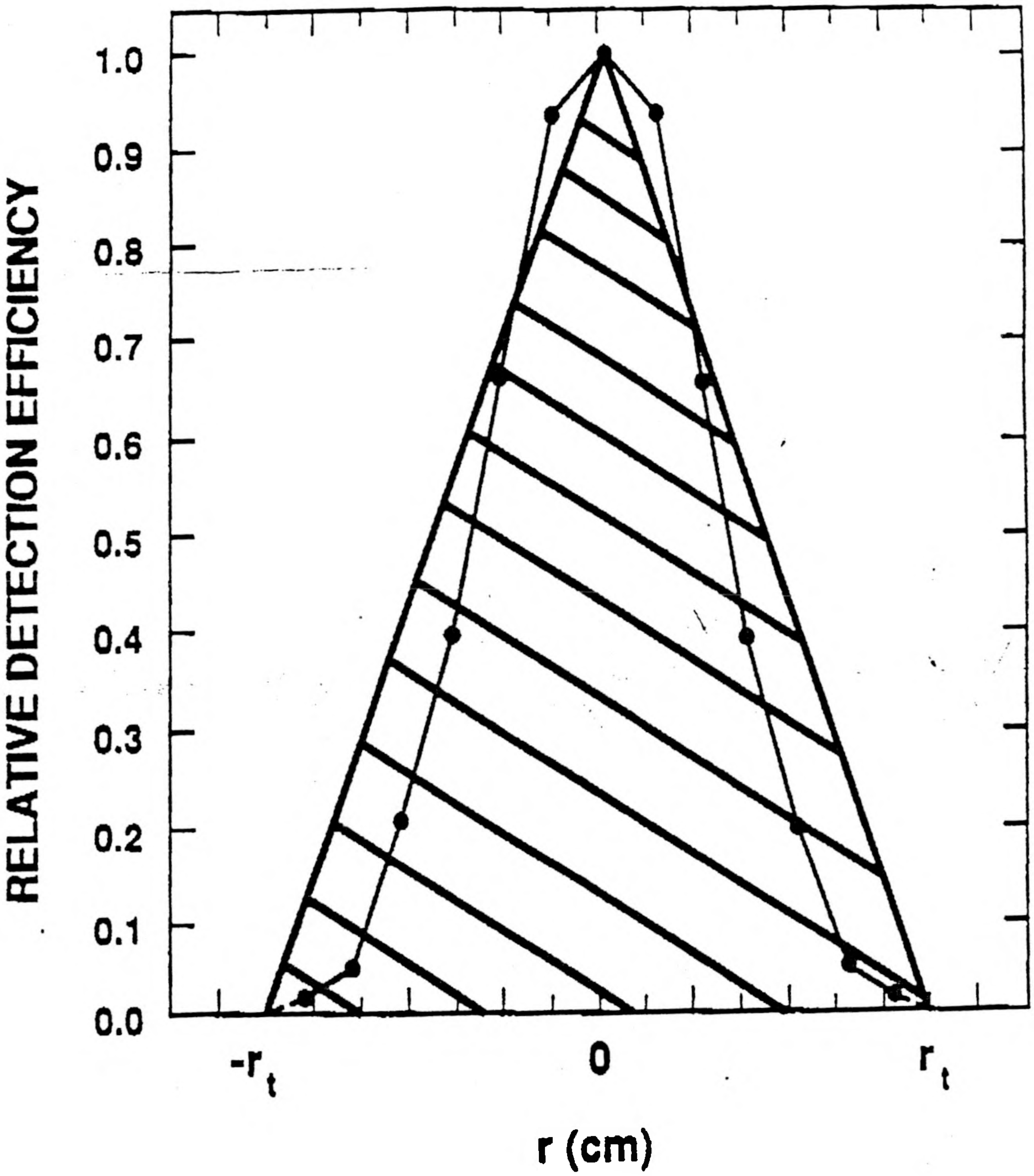
Calibration

- Applied LANL Calibration Technique
- 5 gram Plutonium 'Point Source'
- Correction Factors - Using 'Line Source' Standards
- Correlated to BGO ROI
- Area Calibration Constant Generated



EG&G ROCKY FLATS


r
top
Inner duct
Diameter
W

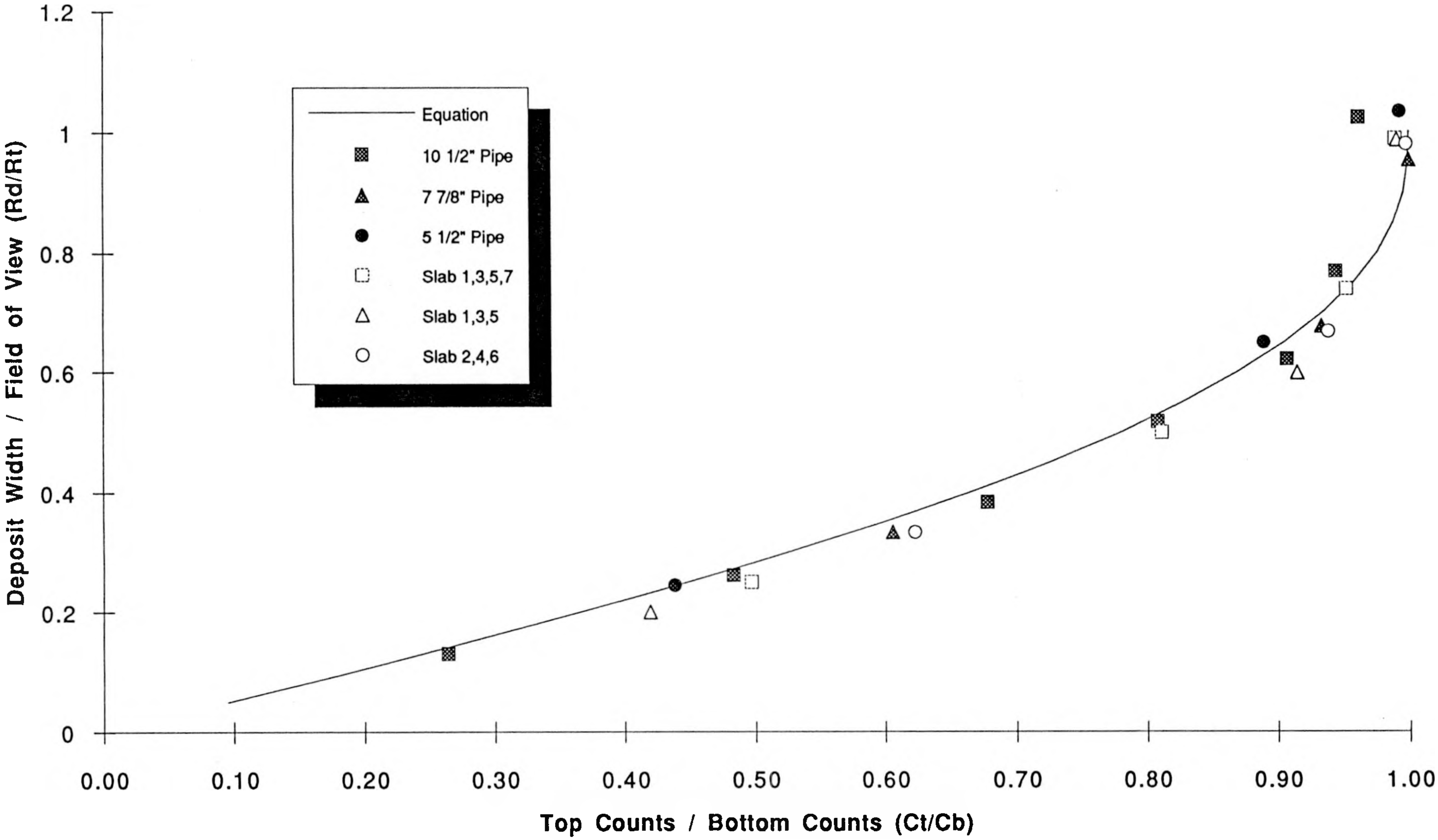

RELATIVE DETECTION EFFICIENCY
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
$-r_t$
0
r_t
r (cm)

$$\frac{C_T}{C_B} = \frac{2}{\Pi}\left[\sin^{-1}(X) + 2X\sqrt{1-X^3} + X^3 \ln\left(\frac{1-\sqrt{1-X^2}}{X}\right)\right]$$

where:

$X = W/2r_{top}$,

$2r_{top}$ = Detector field-of-view projected on duct surface.


1.2
1
0.8
0.6
0.4
0.2
0
Deposit Width / Field of View (Rd/Rt)
Equation
10 1/2" Pipe
7 7/8" Pipe
5 1/2" Pipe
Slab 1,3,5,7
Slab 1,3,5
Slab 2,4,6
0.00
0.10
0.20
0.30
0.40
0.50
0.60
0.70
0.80
0.90
1.00
Top Counts / Bottom Counts (Ct/Cb)

Data Analysis

- Material Width Model
  - Vertical Ducts
  - Top/Bottom Ratio Approaching Unity

- Point Assay (gm/cm) Calculated at Each Location

- Total Pu in Duct Section

Data Acquisition

- Assumption: Majority of Holdup in Bottom of Duct
- Confirmed With
  - Field Measurements
  - Video Characterization
- Measurements Performed Above and Below Duct

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$$PA\ (Gm/Cm) = NCR \times ACC \times W \times CF_{Pipe}$$

(2)

where:

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ACC = Area calibration constant in grams-seconds / cm^2,

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Where:

n = number of assay locations,

y = point assay (gram/cm),

x = position of each point along the length of the duct.

Summary

- Contact Top/Bottom Technique Viable

- Continuing Validation
  - Remediation (Delta)
  - Comparison to 'Far-Field' Method

Duct Clean-out Data

Duct	Duct Length (Ft)	Number of Points Measured	Delta Measurement (Grams Plutonium)	Measured Value (Grams Plutonium)
1	94	125	307	302
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