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Determination of the Radioactive Material and Plutonium Holdup in Ducts and Piping in the 324 Building

**D. L. Haggard
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J. E. Tanner**

January 1996

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest National Laboratory
Operated for the U.S. Department of Energy
by Battelle**



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UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC06-76RLO 1830



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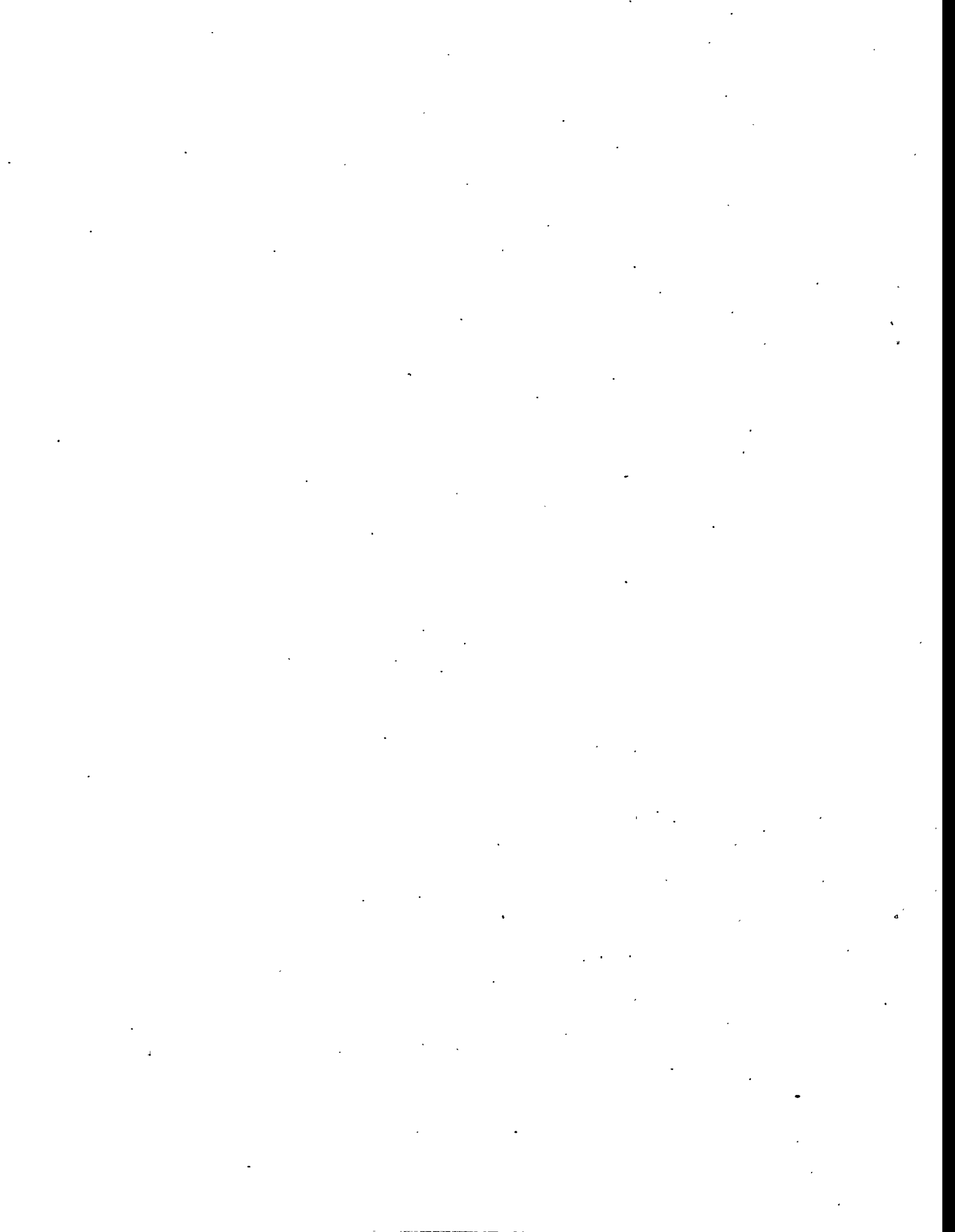
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Pacific Northwest National Laboratory
Richland, Washington 99352

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Executive Summary

This report describes the measurements performed to determine the radionuclide content and mass of plutonium in exposed ducts, filters, and piping in the 324 Building at the U.S. Department of Energy Hanford Site in Washington State. This information is needed to characterize facility radiation levels, to verify compliance with criticality safety specifications, and to allow more accurate nuclear material control using nondestructive assay (NDA) methods. Gamma assay techniques typically employed for NDA analysis were used to determine the gamma-emitting isotopes in the ducts, filters, and piping. Passive neutron counting was selected to estimate the plutonium content because high gamma levels from fission and activation products effectively mask any gamma emissions from plutonium. A high-purity gamma-ray detector was used to measure the mixed fission and activation radionuclides. A neutron slab detector containing five ^3He proportional counters was used to determine the neutron emission rates and estimate the mass of plutonium present. Both measurement systems followed the methods and procedures routinely used for nuclear waste assay and safeguards measurements.

The chronological order of events included a review of previously published documents on holdup assays at other sites and reviews of standards and regulatory guidelines relating to holdup and holdup measurement techniques. Reviews of facility historical radiation survey reports and unusual occurrence reports significantly helped in identifying potential holdup areas. A radiological survey mapping of the pipes and ductwork was performed prior to the actual holdup measurements. During this activity, collection zones were identified and permanently marked, and a unique naming protocol was established. This work was conducted in February 1995.

The fission and activation product inventories in the ducts and piping were measured using gamma-ray NDA techniques. A well-shielded and collimated gamma detector was used to view specific locations on the ducts and piping. The intrinsic germanium detector was used to measure the characteristic gamma-ray spectra of the fission and activation products in the 324 Building. From the gamma-ray intensities measured and the decay scheme of identified radionuclides, it is possible to obtain reasonably accurate measurements of the activity of gamma-emitting nuclides. Almost all the gamma activity present originates from the fission product ^{137}Cs and the activation product ^{60}Co . Radiations from other nuclides present were effectively masked by the intense gamma rays from the mixed fission and activation products. This was particularly true in attempting to directly measure for transuranic (TRU) materials. The small quantities of plutonium present did not produce sufficient gamma activity to be detected in the presence of so much fission and activation product activity. Transuranic activity was determined by measuring neutrons using a ^3He neutron detector. The fast neutrons emitted by plutonium are highly penetrating in metals and can easily be detected, even for gram quantities of plutonium surrounded by several inches of steel shielding.

Special data analysis techniques had to be used to estimate the plutonium content of the ducts since they were not in a standard counting geometry. The mass estimates may be too high if there are a significant number of neutrons produced by alpha-neutron interactions with low-atomic-number

materials in the ducts and pipes, particularly alpha-emitters in intimate contact with fluorine, aluminum, boron, or beryllium. This is of concern if the high-efficiency particulate air (HEPA) filters contain finely divided plutonium oxide powder on borosilicate glass filter media. To be conservative, yet realistic, the neutron detector was calibrated using a known mass of weapons-grade plutonium (6 weight percent ^{240}Pu) in the form of an oxide. This source was well characterized by previous NDA measurements to verify its mass.

The estimates of plutonium mass are highly dependent on the assumptions made in analyzing the data. Specifically, the estimates depend on the isotopic and chemical form of the plutonium, the distribution of plutonium, the amount of intervening shielding surrounding the plutonium, and the general neutron background in the basement and surrounding areas. Assumptions are that the plutonium is in the form of weapons-grade plutonium oxide and at least 10 years old. This is a conservative assumption because much of the plutonium processed in the 324 Building is in the form of high-burn-up material that emits more neutrons per gram (hence, the mass would be less for the same neutron emission rate).

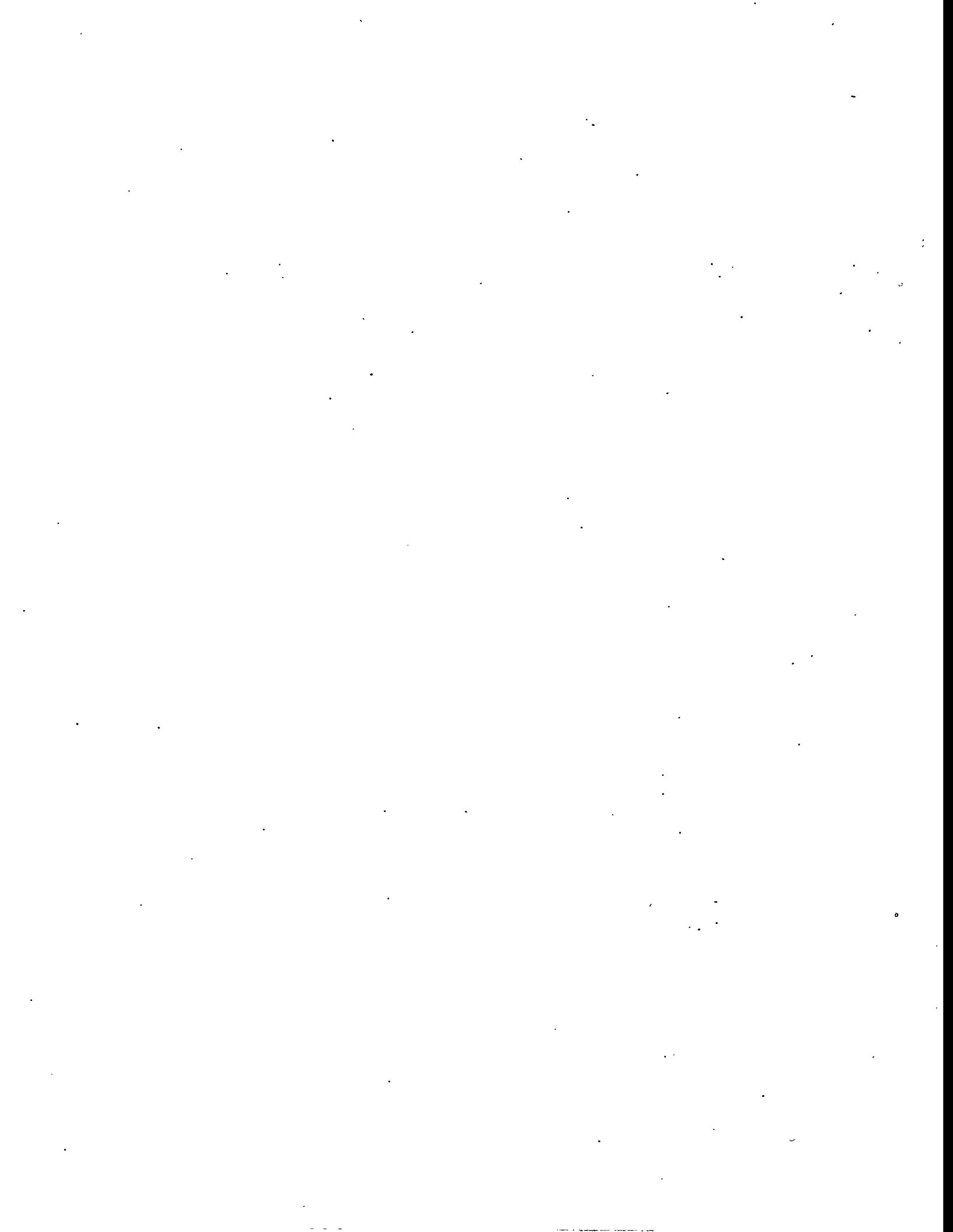
Estimated plutonium mass in the building ductwork and filters is 19.0 grams and the radioactive liquid waste system (RLWS) line has 0.4 grams. Total plutonium mass holdup estimates range from 19.4 grams, assuming that the plutonium is weapons-grade plutonium, to a best estimate of 10.6 grams plutonium, assuming 11% ^{240}Pu .

An estimated 1.19 millicuries of mixed fission and activation products are resident in the ductwork and RLWS piping. Lastly, estimates of the radioactive material in the service gallery (Room 18) were indeterminate due to the high backgrounds and the uncertainty in the locations and distributions of source terms. It appeared that most, if not all, the readings were due to radioactive material in the hot cells and duct space leading to the HEPA filter banks servicing the hot cells.

In summary, the results of this survey indicate no significant levels of plutonium reside in the ductwork or RLWS lines in the 324 Building.

Acronyms

DOE	U.S. Department of Energy
EBR-II	Experimental Breeder Reactor
FFTF	Fast Flux Test Facility
HPGe	high-purity germanium detector
LLNL	Lawrence Livermore National Laboratory
MCA	multichannel analyzer
NDA	nondestructive assay
NIST	National Institute of Standards and Technology
PNNL	Pacific Northwest National Laboratory
RAM	radioactive material
RE	Radiochemical Engineering
RF	radiofrequency
RLWS	radioactive liquid waste system
SMF	Shielded Materials Facility
SNM	special nuclear materials
TRU	transuranic



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1.0 Introduction

This section provides a brief description of the facilities in the 324 Building to provide an overview of radionuclides that may be present in the various cells and ancillary ductwork. The equipment and methods used to provide estimates of the radionuclide inventories are discussed briefly.

1.1 Facility Description

The 324 Building Waste Technology Laboratory is operated by the Pacific Northwest National Laboratory (PNL) for the U.S. Department of Energy (DOE).^(a) The laboratory is used for the study of chemical processes from laboratory to pilot scale and permits the examination and mechanical testing of irradiated specimens. The building contains the Shielded Materials Facility (SMF), the Radiochemical Engineering (RE) cells, the High Bay Engineering Laboratory (HBEL) and the Engineering Development Laboratories (EDL). The 324 Building has a partial basement, first, second, and partial third floors. The outside dimensions of the building are 62.5 m (205 ft) by 71.6 m (235 ft) in plan and 13.7 m (45 ft) in height above the ground.

The primary operating area is on the main floor and includes the SMF and RE hot cells and the operating galleries. Adjacent to the hot cells is a cask-handling area, low-level canyon, manipulator shop, and the truck lock. At the north end of the building is the Support Facilities Addition that serves as the craft maintenance shop for the entire building. The High Bay Engineering Laboratory is a three story structure at the northwest corner of the building that serves as a full-scale engineering development laboratory. The basement contains a fissile material storage vault and Nondestructive Assay (NDA) Laboratory as well as additional laboratories, service tunnel, RE cell service gallery, and tank pit. The second and third floor are mainly offices and supporting laboratories with the third floor also housing the RE cells control room, chemical makeup rooms, and ventilation and equipment rooms. A two-story office addition in the southwest corner houses research and development staff.

The individual hot cells are configured to support a wide variety of operations, and some are maintained free of alpha contamination. Typical work performed in each of the hot cells is described below. These activities may be used as a guide for estimating what types of nuclides and isotopic compositions may be present in the duct work and piping.

The RE cells are located in the north portion of the 324 Building. The RE cell complex consists of A, B, C, D, and Airlock cells. The RE cells and associated service areas provide space for engineering evaluation of processes involving aqueous and solid radioactive and hazardous material. Metallographic investigation of highly radioactive material is also performed. Typical activities include

(a) The Pacific Northwest National Laboratory is operated by the Battelle Memorial Institute for the U.S. Department of Energy under contract DE-AC06-76RLO 1830.

- measuring heat-generating rates of radioactive materials
- characterizing borosilicate glass waste forms for the immobilization of Hanford defense wastes
- repackaging special case wastes not currently accepted for shallow-land burial
- conducting Nondestructive Examination and Destructive Examination on irradiated spent LWR fuels
- treating and packaging small volumes of radioactive and mixed wastes.

The SMF complex is located in the southeast portion of the 324 Building and consists of East, South, and Airlock cells. The SMF is used for detailed evaluation of irradiated fuel and materials, source fabrication, and shipping and receiving radioactive materials. Some of the activities include:

- gamma scanning on CsCl capsules, Fast Flux Test Facility (FFTF) fuel pins; EBR-II fuel pins, and SP-100 fuel pins
- radiography of encapsulated radioactive materials, e.g., gamma radiation or heat sources
- postirradiation examinations on tritium-bearing components and experiments, including sectioning/partitioning, gas recovery, and sample analysis.

1.2 Radiological Survey

Holdup measurements are required in all facilities where special nuclear materials (SNM) may exist in building systems. Potential criticality safety issues have been identified at other DOE facilities and have been addressed in PNL-MA-25, *Criticality Safety*, as action items.^(a) The major objective of this project is to identify and quantify radioactive materials that may contain transuranic (TRU) holdup in various ducts, processes, and equipment in the 324 Building. The radiological mapping survey was used 1) to locate potential sources of radioactive materials/special nuclear materials (RAM/SNM) at specified locations; 2) to provide a baseline reference point for trend analysis on data from future measurements; 3) to augment ongoing radiological surveys; and 4) to provide supporting documentation regarding criticality safety and provide data for nuclear material accountability.

A radiological survey of all 324 Building cell ductwork, radioactive liquid waste system (RLWS) lines, and laboratories in the basement was conducted in February 1995. The survey readings were taken by PNL radiological control technicians. Guidelines for holdup assay are stated in U.S. Nuclear Regulatory Commission (NRC) Regulatory Guides 5.23 and 5.37 (1984, 1983) and the Los Alamos Nuclear Safeguards Training Program (LANL 1992).

(a) Pacific Northwest Laboratory (PNL). August 17, 1994. *Criticality Safety*. Controlled technical manual, PNL-MA-25. Pacific Northwest Laboratory, Richland, Washington.

1.2.1 Radiological Survey Equipment

Equipment used for the survey included a gieger-mueller (GM) counter, a Cutie Pie, and a marking device for permanently labelling measurement locations. The potential hazards identified were possible contamination and high-dose-rate areas. Personnel safety issues were addressed and covered in the radiological work procedure specific to this work. The radiological survey procedure followed regulatory guidelines. Preliminary radiation survey measurements of the facility were used to budget the measurement time, to emphasize high-holdup areas, to establish independent collection zones, and to determine detector positions within the zones.

1.2.2 Delineation of Collection Zones and Assay Sites Procedure

The procedure used to establish assay locations is given below.

1. In each room suspected of having material holdup, detector positions (assay sites) were chosen so that the material holdup could be measured with both the neutron slab detector and the HPGe detector.

Each room usually contained several types of holdup areas. For example, Room 3K contained basement ductwork, old ductwork for a dismantled glovebox, and a fume hood with its associated flue and ductwork.

2. Each assay site was marked with a permanent marker to ensure reproducible assay positions.

For ducts and RLWS pipes, dose rate measurements were taken at successive 2-ft intervals, beginning 1 ft in from the start of the duct or pipe. Where dose rates remained constant over long sections of piping or ductwork, measurements were spaced out over larger distances but usually multiples of 2 ft. Interval points were marked and identified using permanent ink or equivalent.

"Hot spots" were also identified, surveyed, and marked. Dose rate information was recorded on a data sheet provided by the task leader.

3. Each site was uniquely labeled to facilitate unambiguous reference to that site in the assay log. A labeling convention was established.

Labeling proceeded sequentially for each room, beginning with room number/letter designation followed by the interval identification number. For example, the location of the first measurement point in room 3K was 3K_1. Measurements began in the basement of the 324 Building and progressed to the first floor areas.

1.3 Gamma Assay

Gamma-ray assay is based on the activity observed from specific energy peaks detected from the isotopes present. Equipment for gamma holdup measurements consisted of the following components:

- tungsten collimator
- high-purity germanium (HPGe) detector - efficiency 35%
- angular response of the detector/collimator reference (see Figures 1.1 and 1.2)
- Canberra Genie-PC AIM Data Analysis and Acquisition System.

The HPGe detector was shielded and collimated from other surrounding source material. A tungsten collimator was used with the gamma-ray detector. Additional shielding was also needed to eliminate background gamma rays originating behind the detector. The collimator provided sufficient shielding and directionality for the holdup measurement geometry. The detector's field of view was defined by the tungsten collimator. Calibration of the gamma system was performed with the same collimation setup used during the holdup measurement campaign.

Initial calibrations for the holdup measurement system were performed for point, line, and area source geometries. These point, line, and area geometry calibration constants described the distribution in the detector's field of view. The correction factors were specific to the detector and collimation

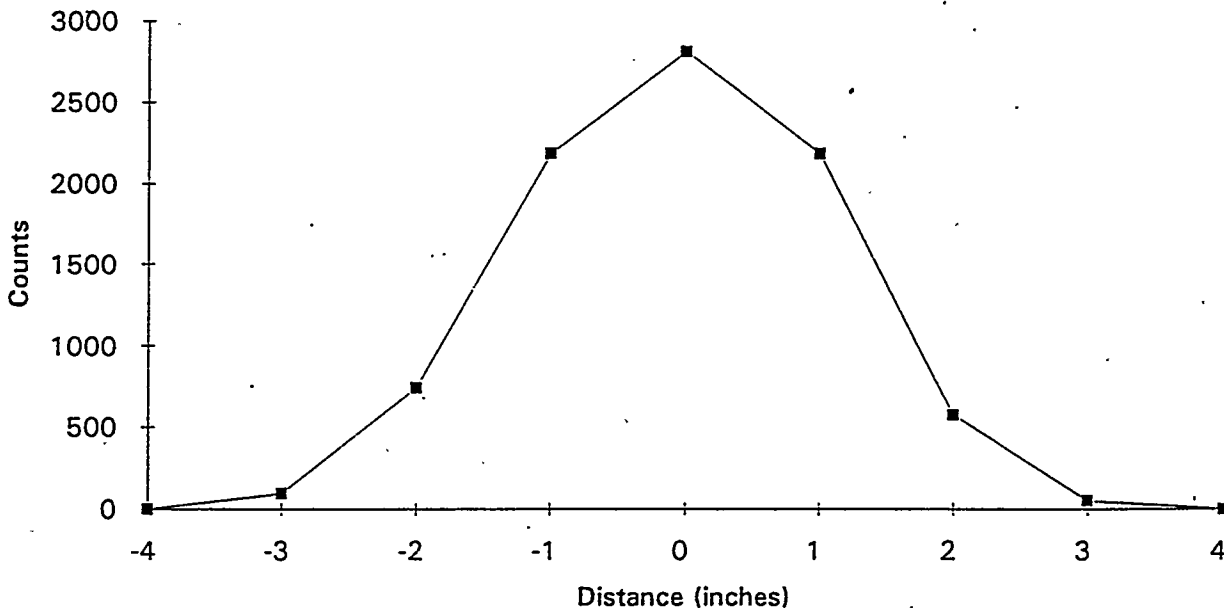


Figure 1.1. Point Source Measurements by Collimated High-Purity Germanium Detector at Selected Distances from Source

327 COLLIMATOR #1
12" FROM SOURCE

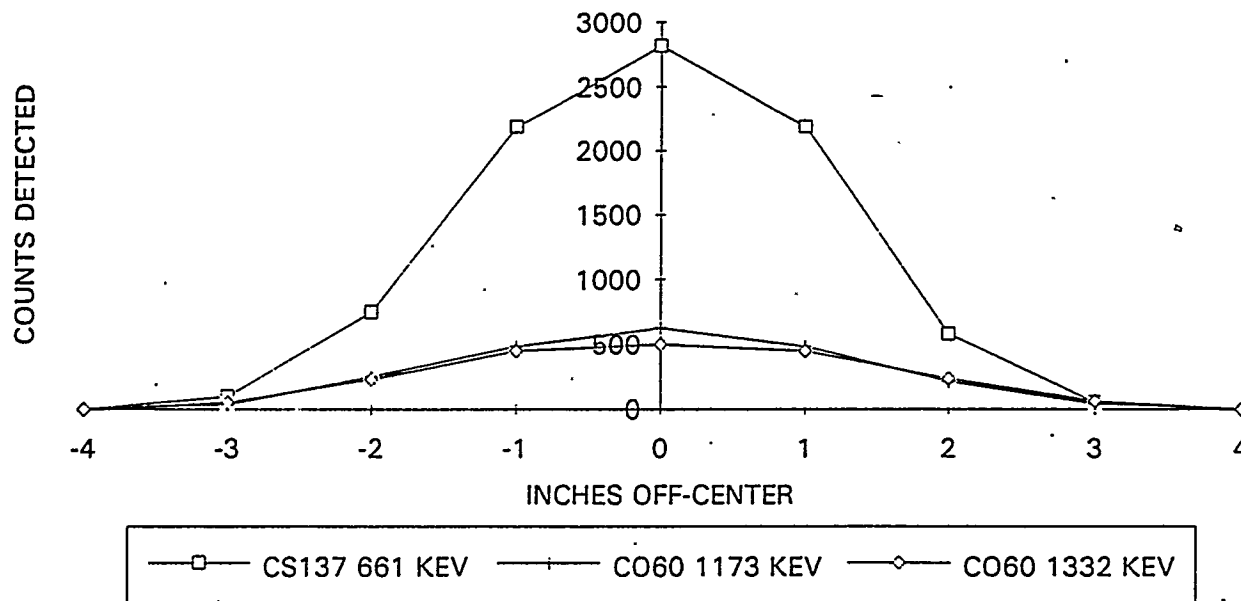


Figure 1.2. Distribution of Gamma-Ray Activity Along Horizontal Measurement Axis of the HPGe Detector

setup and were used to correct for the detector's radial response to the incident radiation of interest. These corrections were used in the correlating measurements to convert the results to activity or grams of material. This procedure used a single point source to determine these correction factors. Point source measurements were conducted at several horizontal positions, as indicated in Figure 1.1. Figure 1.2 illustrates the gamma detector's response along the horizontal measurement axis. The gamma detector's relative efficiency for that geometry was determined. One reference standard centered in front of the detector at a fixed distance was measured to determine the detector's absolute response to the incident radiation.

Formulae required to determine calibration constants are as follows:

$$L = 2s \frac{\sum_{i=0}^n C_i}{C_0} - s \tag{1.1}$$

- where L = effective length of the detector field of view at distance r_0
- s = length of the horizontal increment
- C_i = net count rate at horizontal position i
- C_0 = net count rate with the source centered in front of the detector.

$$a_0 = \frac{\pi s^2}{4} \quad (1.2)$$

where a_0 is the incremental area at horizontal position 0

$$a_i = \pi [(i + 0.5)s]^2 - \pi [(i - 0.5)s]^2 \quad (1.3)$$

where a_i is the incremental area at horizontal position i

$$A = \frac{\sum_{i=0}^n (a_i C_i)}{C_0} \quad (1.4)$$

where A is the effective area of the detector field of view at distance r_0

$$K_p = \frac{m_0}{C_0 r_0^2} \quad (1.5)$$

$$K_l = \frac{m_0}{LC_0 r_0} \quad (1.6)$$

$$K_a = \frac{m_0}{AC_0} \quad (1.7)$$

where K_p = point source calibration constant ($\text{g}\cdot\text{s}/\text{cts}\cdot\text{cm}^2$)

K_l = line source calibration constant ($\text{g}\cdot\text{s}/\text{cts}\cdot\text{cm}^2$)

K_a = area source calibration constant ($\text{g}\cdot\text{s}/\text{cts}\cdot\text{cm}^2$)

m_0 = mass of the reference source (g)

C_0 = net count rate of the reference standard centered in front of the detector face at distance r_0 (cts/s).

Once calibration constants are determined, quantitative estimates can be made for point sources, line sources, and area sources.

1.3.1 Point Source Calibrations

A point source is a small deposit centered in a large detector field-of-view. Its calibration is determined with Equation 1.8:

$$m = K_p C r^2 \quad (1.8)$$

where m = mass of isotope of interest (g)
 K_p = point source calibration constant (g·s/cts·cm²)
 C = net count rate (cts/s)
 r = source-to-detector distance (cm).

1.3.2 Line Source Calibrations

A line source is a narrow uniform deposit, centered in a wide detector field-of-view, which spans the width of the detector field-of-view. Its calibration is found with Equation 1.9:

$$m = K_l C r l \quad (1.9)$$

where m = mass of isotope of interest (g)
 K_l = line source calibration constant (g·s/cts·cm²)
 C = net count rate (cts/s)
 r = source-to-detector distance (cm)
 l = length of a line source (cm).

1.3.3 Area Source Calibrations

An area source is a uniform deposit that fills the detector field-of-view. Its calibration is found with Equation 1.10:

$$m = K_a C a \quad (1.10)$$

where m = mass of the isotope of interest (g)
 K_a = area source calibration constant (g·s/cts·cm²)
 C = net count rate (cts/s)
 a = area of an area source (cm²).

The equipment was moved to the 324 Building and checked for physical damage. A combination standard traceable to the National Institute for Standards and Technology (NIST) was measured periodically during the measurement campaign to demonstrate that the instrumentation was properly functioning and that the calibrations determined in the laboratory were still valid.

1.4 Neutron Assay

Accurate determination of the fissile material and radioisotope content in the ducts, filters, and piping of the 324 Building is a very difficult problem. Nondestructive assay techniques for fissile materials usually employ gamma spectroscopy to identify and quantify each gamma-emitting nuclide, such as plutonium. However, the massive amounts of fission products present completely obscure any gamma rays emitted by plutonium. Fortunately, the neutrons emitted by plutonium are highly

penetrating and can easily pass through several inches of the steel or lead shielding used to reduce gamma doses, although the neutrons may be degraded in energy. With some degree of uncertainty, the amount of plutonium can be determined using appropriate calibration standards and neutron detectors. Passive neutron counting is a recognized technique for nondestructive assay of plutonium and is described in Chapter 14, "Principles of Neutron Counting" in *Passive Nondestructive Assay of Nuclear Materials*, NUREG/CR-5550 (Reilly et al. 1991).

Neutrons emitted by plutonium originate from two main sources:

- spontaneous fission of even-numbered plutonium isotopes
- alpha-neutron reactions with any low-atomic-number materials in intimate contact with the plutonium.

Most of the neutrons originate from ^{238}Pu , ^{240}Pu , and ^{241}Am , which is the decay product of the short-lived nuclide ^{241}Pu . Thus, it is important to know not only the isotopic composition of the plutonium, but also the time since chemical separation or the ^{241}Am content to be able to estimate the neutron contribution from ^{241}Am . In many practical situations, the number of neutrons produced by alpha-neutron reactions can exceed the spontaneous fission production from plutonium. Any alpha-emitter in contact with low-atomic-number nuclides will produce neutrons. For example, transuranics that are in intimate contact with beryllium, boron, aluminum, sodium, or even concrete will produce neutrons. High neutron emission rates can result from finely divided plutonium oxide dust on borosilicate glass or aluminum spacers in HEPA filters. The neutron emission rates can be tens to hundreds of times higher than pure plutonium metal.

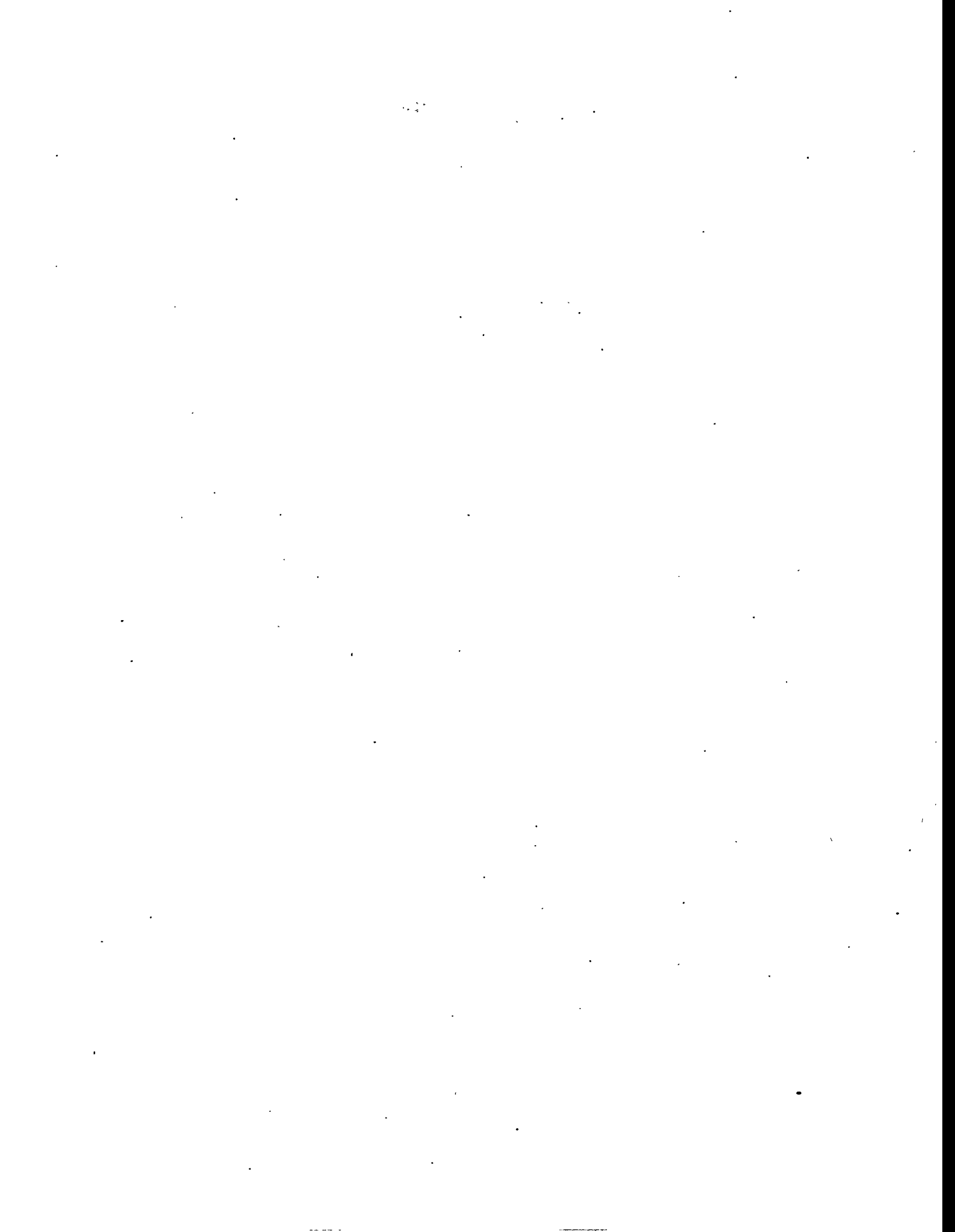
It is conceivable, although not likely, that the material may not contain any plutonium, but will still emit neutrons from alpha-neutron reactions from americium or other transuranic material with high alpha activity. The exact amount of plutonium present can be determined from the neutron measurements if we know the following:

- the exact isotopic composition of the plutonium
- the time since chemical separation to allow calculation of ^{241}Am progeny
- chemical composition of the plutonium
- presence of low-atomic-number impurities in contact with alpha-emitters
- other transuranics present that may also emit neutrons, such as ^{244}Cm .

We can make some simplifying assumptions that will limit the range of calculated values of the amount of plutonium that may be present. For instance, we can assume that the plutonium will be low-exposure plutonium. Neutron emission rates of weapons grade (less than 6% ^{240}Pu) or N-Reactor

plutonium are quite low, typically about 150 neutrons per second per gram of plutonium. Higher-exposure plutonium would emit more neutrons and result in a smaller actual mass for a given measured neutron emission rate.

Plutonium metal is chemically unstable, and any small pieces would eventually convert into an oxide or oxide-hydroxide mixture in moist air. The oxide is the most chemically stable form of plutonium and would be the most likely form to be found. The neutron emission rates of oxylates, nitrates, and hydroxide mixtures will be somewhat similar to that from pure plutonium oxide. Assuming any plutonium present would be in the form of an oxide will result in conservative estimations. Any other chemical form, particularly fluorides, would emit more neutrons and result in a smaller actual mass for a given measured neutron emission rate. However, oxides are the most probable chemical form found in the reactor fuel.



2.0 Scope and Objectives

The objectives of this work are threefold:

- provide a baseline radiological survey of the ducts and piping
- determine an upper limit of plutonium inventory and an estimate of the most probable amount of plutonium that may be present
- locate and quantify the amount of fission, activation, and TRU materials that are present in ducts, RLWS lines and other facility processes, as a baseline for future reference.

This work is limited in scope to the nondestructive assay of the contents of the ducts, filters, and piping in the 324 Building and low-level waste that resided in those locations at the time measurements were taken.

2.1 Definition of the Problem

Over many years of operation of the hot cells in the 324 Building, the internal ducts, filter boxes, and piping and drain lines from the hot cells have become contaminated with large amounts of fission and activation products and small amounts of plutonium and other transuranic nuclides. The fission and activation product inventory is large enough in some locations that it presents a radiation hazard, and in some locations steel and lead shielding have been added to critical areas to reduce radiation doses. It is conceivable that enough plutonium and transuranic material has accumulated to be of concern for criticality safety and safeguards controls.

The purpose of this work is to attempt to quantify the amounts of radionuclides in the internal ducts, filter boxes, and pipes leading from the hot cells. Of particular concern are the liquid waste lines that may have accumulated fuel debris, with the potential build-up of quantities of plutonium. Because of its high activity, the radioactive cesium and cobalt are relatively easy to locate. However, the high gamma levels will mask the gamma emissions from plutonium and other transuranics. The problem is further exacerbated by the streaming of neutrons down ducts and openings from the hot cells. Inventories of plutonium, curium, and other neutron-emitting nuclides in the hot cells will create variable backgrounds in the basement in the area around the ducts and pipes. It is extremely difficult to accurately assess plutonium inventories in the presence of variable neutron backgrounds. Passive neutron measurements are further complicated by the storage of over 70 grams of ^{238}Pu oxide stored in the vault in the basement of the 324 Building. The neutron emission rate from the ^{238}Pu oxide is orders of magnitude higher than from other sources we are attempting to measure.

2.2 Planning and Solution

A review of the radiological conditions was made, including gamma surveys to determine hot spots in the ducts and piping. Excessive gamma dose rates from fission products preclude the use of gamma assay equipment routinely used to determine plutonium mass. The high dose rates from fission products would interfere with the plutonium isotopic analysis. Therefore, neutron assay techniques would be more reliable for locating and quantifying fissile material. A neutron slab detector based on a design from the Los Alamos National Laboratory (LLNL) Safeguards Assay Group was selected for measurements to determine the amount of plutonium that could be present. The amount of plutonium in the ducts and RLWS pipes can be estimated by:

- calibrating the neutron detector with appropriate calibration standards
- measuring the neutron flux around the ducts, filter boxes, and piping
- carefully analyzing the data to relate the measured neutron fluxes to mass of plutonium.

The gamma-emitting nuclides were readily identified by standard gamma-assay techniques because in most instances the nuclides are not heavily shielded in the ducts and pipes. In most situations, the amount of shielding is well known and the radioactive material localized. A well collimated intrinsic germanium detector was used to quantify gamma-emitters. Where space permits, measurements are made at more than one orientation and the results averaged to give an accurate estimate of the contents inside ducts and pipes.

3.0 Measurements

The measurements were conducted according to the following documents:

- Pacific Northwest Laboratory Quality Assurance Plan QAP FO-1, *QA Plan for Safeguards Nondestructive Assay Measurements*
- Pacific Northwest Laboratory Nondestructive Assay Measurement Procedure MCA-510, *Holdup Assay Measurements* (see Appendix A).

3.1 Measurement and Test Equipment

The neutron and gamma assay systems are "user to calibrate" equipment. The individual components are configured and tested as a system, and then the entire system is calibrated with appropriate calibration/verification standards. The neutron and gamma assay systems are calibrated with gamma check sources with calibrations standards traceable to the NIST.

3.2 Calibration Standards

Calibration standards are used in a measurement system to establish the relationship between the basic instrument response and the attributes of interest. The quality of the calibration is ensured by selecting the appropriate standard. The standard must be a physically and chemically stable item for which the attributes of interest are well characterized and for which other properties affecting the measurement are known. In this case, the count rate from neutrons, the amount of shielding, and the orientation and distance from the detector to the neutron source are related to the mass of weapons grade plutonium. The mass of other forms of plutonium can be inferred from the calculated neutron emission rates. The calibration of the gamma assay equipment is more straightforward. The counting efficiency of the gamma spectrometer with a collimator can be measured using calibration sources of known activity and photon emission rates. The viewing angle and efficiency can be determined directly from measurements, as shown in Figure 1.2.

Records indicate that the material in the hot cells was from a wide variety of sources, including both low-exposure and high-exposure reactor fuel samples. For these measurements, it was assumed that the plutonium would be low-exposure plutonium oxide with relatively low neutron emission rates (typically 150 neutrons/second/gram of plutonium). To give conservative results, any high neutron yields from alpha-neutron reactions were ignored. The neutron energy spectrum from plutonium dioxide is shown in Figure 3.1, taken from the document *Neutron Spectra of Plutonium Compounds*, BNW-1262 (Brackenbush and Faust 1970).

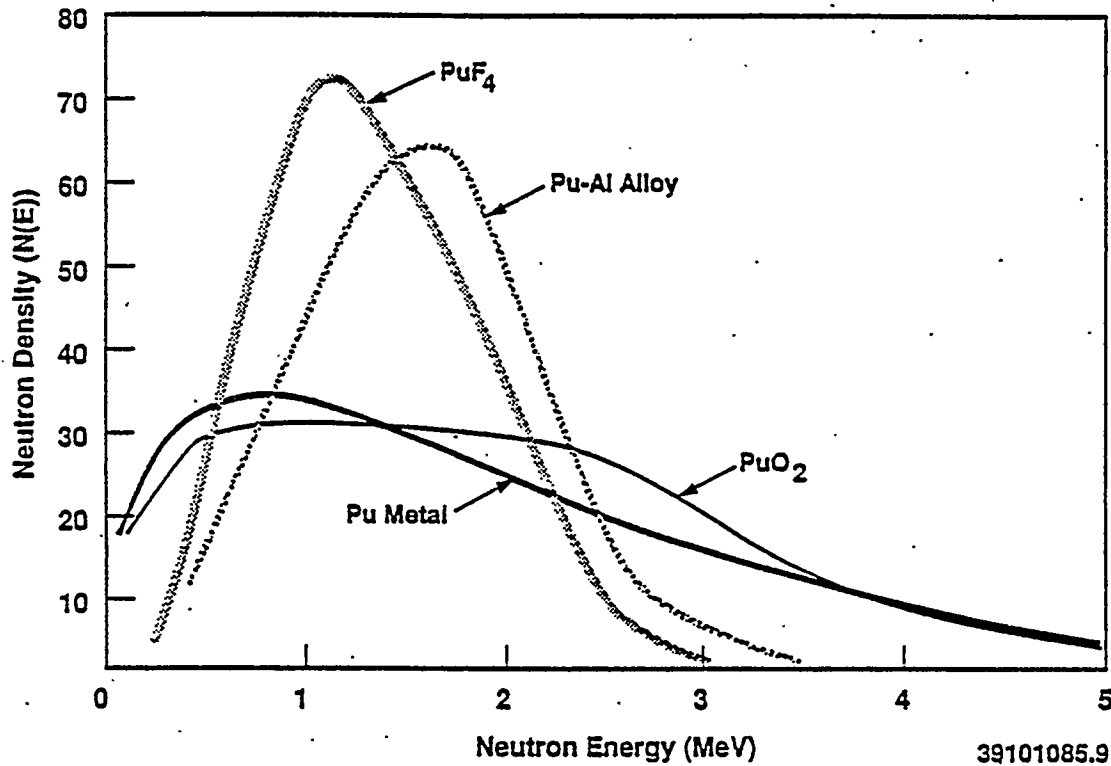


Figure 3.1. Neutron Energy Spectra from Plutonium Compounds

Two neutron sources were selected for use in calibrating/verifying the neutron detector assay system. The first was a small $0.3\text{-}\mu\text{g}$ ^{252}Cf neutron calibration source. This source's neutron dose rates have been determined by comparison with other neutron sources whose calibrations are directly traceable to NIST. This source has a neutron energy spectrum very similar to that of plutonium dioxide. (See *^{252}Cf Shielding Guide*, DP-1246 [Stoddard and Hootman 1971] or NBS Special Publication 633 [Schwartz and Eisenhauer 1982].) This source was used to determine the angular efficiency of the slab detector in the laboratory prior to the holdup measurements at the 324 Building.

A sample of weapons-grade plutonium oxide was selected as the reference material standard for determining the basic instrument response for the attributes of interest. Specifically, the mass of plutonium in the reference material was related to the neutron count rate in the neutron detector. Weapons grade plutonium has a lower neutron emission rate than the higher exposure plutonium samples in the hot cells and should yield conservative (i.e., higher) plutonium mass estimates. The plutonium oxide sample (LLNL ash sample RA 146A) selected for the reference standard contains 108 grams of plutonium with the isotopic composition given in Table 3.1. The reference standard has been measured by several nondestructive assay (NDA) systems used for measuring safeguard mass

Table 3.1. Isotopic Composition of Plutonium Ash Reference Material, Weight Percent Isotope

	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu	²⁴¹ Am
Weight Percent Isotope	0.13825	93.452	6.101	0.2793	0.019	0.220
	± 1.53%	± 0.15%	± 2.21%	± 1.22%	n/a	± 1.4%

Calculated age from ratio of ²⁴¹Am/²⁴¹Pu is 12.3 ± 0.33 years.

confirmatory standards. Data for this sample from high-level neutron coincidence counting and calorimetry with isotopic compositions obtained from gamma spectroscopy are in good agreement with the book values assigned by LLNL when the material was shipped to PNL.

3.3 Measurement and Quality Control

The measurement personnel had the complete responsibility for monitoring and evaluating the quality of the data. Any indication that the system was out of calibration (e.g., significant gain shifts in the position of the thermal neutron peak or high gamma levels that produce pile-up in the neutron events) required the stoppage of work until the problem was resolved. Before NDA measurements were initiated, the overall system was calibrated. To verify the proper operation of the neutron assay system, the entire system was assembled in the laboratory and components checked for proper operation. Then, verification measurements were performed using a known mass of plutonium before and after the measurements in the 324 Building. All of the spectral data collected during the measurements were recorded on computer files and can be retrieved if questions should arise.

A measurement control check was performed daily before the equipment was used, and the results recorded. For the neutron assay system, the detector was positioned at a fixed location 12 in. from a ²⁵²Cf reference source stored in the 324 Building vault. The measurement control check was used to check the position of the thermal neutron peak and verify that the integral of the neutron events for a 200-second count remained constant throughout the duration of the measurements. For the gamma assay system, the germanium detector was exposed to a calibration source containing a known activity of radioactive material. An analysis of photopeak areas and channel number is used to verify that the gain and sensitivity has remained constant. These procedures are recorded in Appendix A, and the data are archived on magnetic storage media.

3.3.1 Preparations for Measurement and Test Equipment

The approach to the problem of NDA assay was to select a very sensitive neutron detector to measure the neutrons emitted by any TRU (plutonium) contained in the ducts, filter boxes, and pipes. The detector requirements included sufficient sensitivity to measure sub-gram quantities of plutonium at accessible measurement locations. A slab neutron detector was selected for the measurements because of its sensitivity and directionality.

3.3.2 Neutron Slab Detector System

The neutron detector system consists of

- a neutron slab detector containing several ^3He proportional counters inside a polyethylene moderator
- ancillary NIMbin electronics (preamplifier, shaping amplifier, high voltage power supply)
- a multichannel analyzer (MCA) to display and record the neutron spectra from the detector.

The neutron slab detector consists of an array of five cylindrical ^3He proportional counters, 1 in. in diameter by approximately 22 in. long, which are filled with four atmospheres of ^3He to detect slow neutrons. The proportional counters are inside a slab of polyethylene moderator at a depth of 2 in. from the front face and 4 in. from the back face. The tubes are carefully selected to have the same gain and are connected in parallel to a preamplifier and high voltage supply. The nominal operating voltage is +1400 volts. The polyethylene slab is 6 in. deep, 16 in. wide, and 24 in. long. The detector is a standard design used for NDA safeguards assay and is described in Section 14.4.2 of NUREG/CR-5550, *Passive Nondestructive Assay of Nuclear Materials* (Reilly et al. 1991).

The NIMbin electronics include a high-voltage power supply to provide a regulated voltage of +1400 volts DC to operate the proportional counters and a shaping amplifier to convert the signals from the preamplifier to pulses that can be processed by the MCA. An MCA was used to display the spectra, so that any possible gamma interference or instrument malfunction could be detected immediately. Data from the MCA (Canberra Series 35 Plus) were recorded on a lap top computer with a hard disk for permanent data storage. Data from the computer was also backed up on floppy disks as a precaution. If there is any question about a measurement, the spectral data can be retrieved and examined later.

3.3.3 Gamma Assay System

Two HPGe detectors were used to determine radionuclide identification and quantities present at various measurement locations. One was a 4.2% efficient detector. It was used to measure the higher-dose-rate areas which could not be measured by the other 35% efficient detector due to high count rate and pulse pile-up. Some locations were measured with both detectors. The results of these

measurements were compared and used to provide an error estimate at the measured location. The gamma assay system is composed of the latest innovative hardware and software commercially available.

3.4 Pretest Verifications

Pretest verifications consisted of testing the electronic equipment, making angular response measurements, and calibrating the slab detector with plutonium oxide.

3.4.1 Testing of Electronics

The neutron slab detector and supporting electronics were assembled in the laboratory, and all of the electronic components were checked to assure proper operation in the field. The electronics were placed in the NIMbin and the cables connected for the signals, high voltage, and preamplifier power. The methods used to verify the proper operation of the equipment in the laboratory generally followed the methodology given in *Neutron Dosimetry at Commercial Nuclear Plants*, NUREG/CR-3610 (Brackenbush et al. 1984), which describes the setup of neutron spectrometry equipment using ^3He proportional counters. The output signals from the shaping amplifier was examined by exposing the detector to a ^{252}Cf neutron source in the laboratory and observing the pulses with an oscilloscope to examine the pulse shape. Adjustments are made to correct for amplifier gain and pole zero to minimize pulse overshoot or undershoot, so that the pulse returns to the baseline in the shortest possible time to reduce pulse pile-up and allow operation in high count rates. Because this adjustment depends on the cable capacitance, the entire system was tested, including the long cables that were to be used. If any component failed, there were backup units that were also checked.

The equipment was calibrated as a complete system before the 324 Building measurements were initiated. Following safeguards measurement procedures, the system was calibrated at the system level. It is not necessary, or even desirable, to calibrate the individual components of the system, because proper operation of the system and accurate interpretation of the results cannot be obtained from individual component electronic calibration. System malfunctions usually can be easily identified from the spectra recorded during the measurements. Proper operation of the system was verified with measurements using a known mass of plutonium.

The calibration/verification measurements consisted of measuring the angular response and response to room-scattered neutrons using the 0.3- μg ^{252}Cf neutron source in the ESB Building at PNL. The response of the detector would then be known at any angle and distance to a fission source. The mass of the plutonium was then correlated to the neutron count rate from the slab detector by exposing the detector to the 108-g plutonium oxide source in the 324 Building.

Figure 3.2 shows a typical spectrum obtained from the ^3He proportional counters in the slab detector exposed to a neutron source. In this graph, the ordinate is the logarithm of the number of counts from the detector, and the abscissa is the energy deposited in the detector by gamma rays and neutrons.

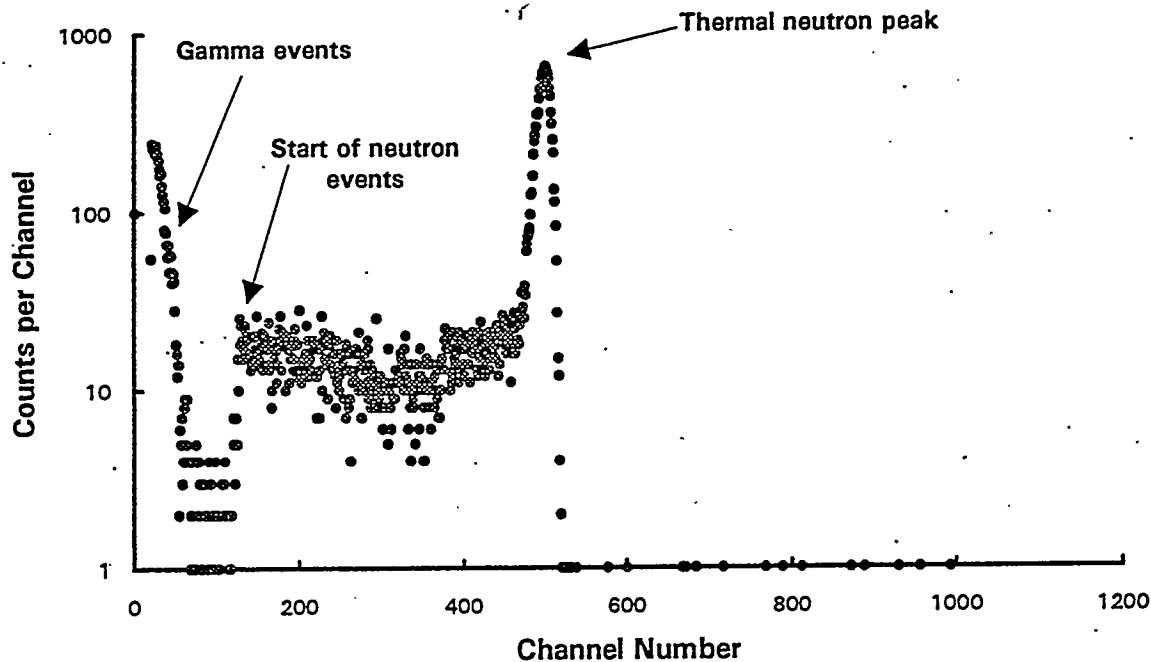


Figure 3.2. Energy Deposition Spectrum from the ^3He Proportional Counters Exposed to a Neutron Source

There is a clear separation between gamma events, shown on the left side of the plot, and neutron events. Gamma events can be eliminated by setting a region of interest and integrating only neutron events as shown in Figure 3.2.

However, pulse pileup can occur in intense gamma fields, when several gamma pulses are counted within the resolving time of the electronics. This produces a gamma continuum that can extend into the neutron event region. The slab detector was exposed to a ^{137}Cs gamma source in the ESB Building laboratory to determine the operating range in high radiation fields. There is no significant gamma pile-up that could interfere with the neutron detector at gamma exposure rates up to 1 R/h. The detector can function in gamma fields as high as 35 R/h, but there is serious gamma pile-up, and the region of interest must be adjusted. The spectral data were recorded, so that all of the measurements can be reviewed later, if necessary.

3.4.2 Angular Response Measurements

The first set of calibration/verification measurements were made in the ESB Building laboratory, using the $0.3\text{-}\mu\text{g } ^{252}\text{Cf}$ neutron source to set up the system electronics and determine the angular response of the slab detector. The response of the detector as a function of polar and azimuthal angles is given in Figures 3.3 and 3.4, respectively. Angles are measured in reference to an axis normal to the center of the front face of the slab detector. (The front face has the five ^3He tubes located at 2 in. below the surface of the plastic moderator.) The azimuthal angles are measured in the plane passing

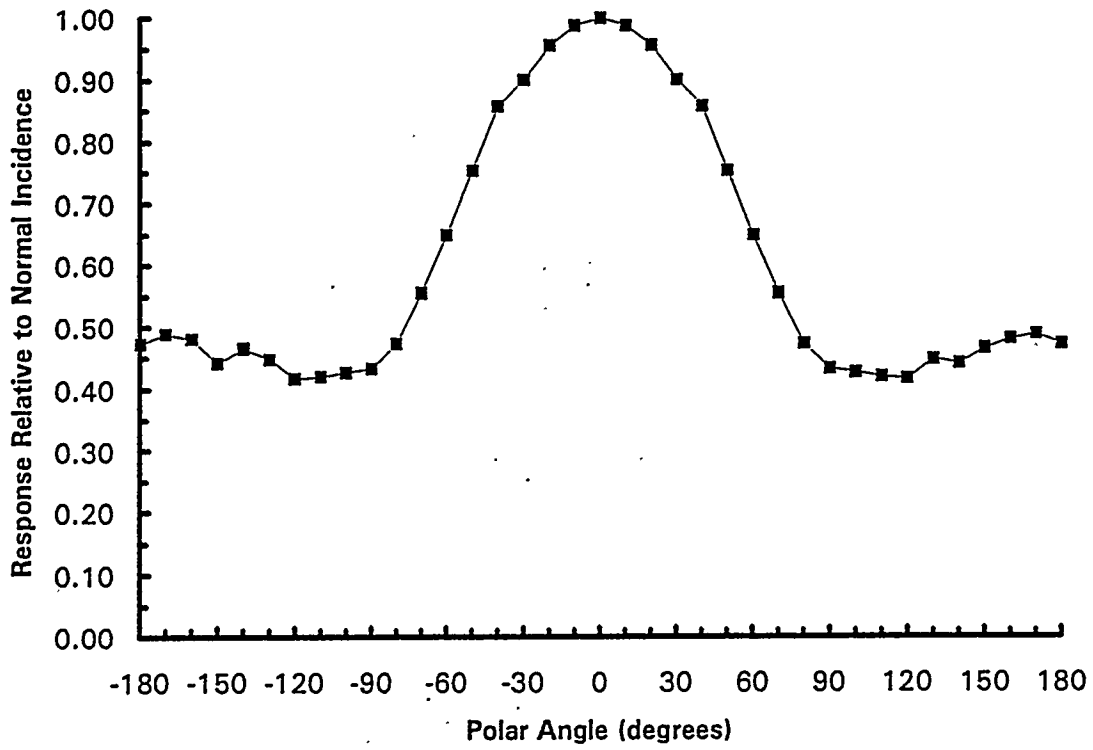


Figure 3.3. Relative Angular Response of the Slab Neutron Detector as a Function of Polar Angle

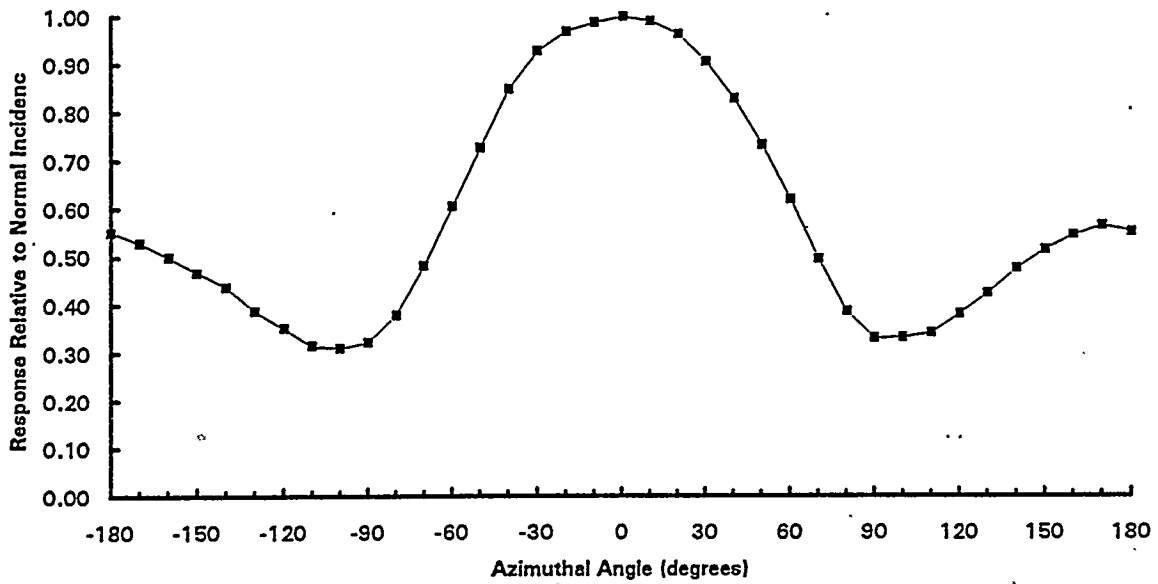


Figure 3.4. Relative Angular Response of the Slab Neutron Detector as a Function of Azimuthal Angle

through the longest axis of the rectangular slab; polar angles are measured in a plane at right angles to the longest axis of the rectangular slab. These measurements were made in the ESB Building laboratory at a height of 4 ft above the floor. The angular responses were measured with the slab detector positioned vertically on a turntable; the neutron source was at a distance of 6 ft (2 m) from the center of the ^3He tubes. Details of the measurements are given in Appendix B. The graphs demonstrate that the angular counting efficiency does not change dramatically if the source is slightly off-axis, and that neutrons entering the back side of the detector are counted at about 50% of the efficiency of those entering the front.

3.4.3 Calibration of Slab Detector with Plutonium Oxide

The slab detector was also calibrated in the 324 Building by exposing the detector to a 108-g sample of plutonium oxide (LLNL sample RA 146A) at a height of 4 ft above the floor and in a vertical configuration. The 108-g plutonium source has been described previously in Section 2.0. As will be explained in Section 4.0 on data analysis, it is possible to correct for the effects of room-scattered neutrons from the series of measurements that relate distance to the measured neutron count rate. After these measurements were completed, it was possible to relate the mass of weapons-grade plutonium to the measured count rate, the distance from the detector, and the angle of incidence. As will be explained in the data analysis in Section 4.0, it is even possible to correct for the effects of neutrons scattered within the facility from inverse square measurements performed in the 324 Building.

Measurements were also made in the 324 Building using various thicknesses of steel and aluminum to estimate the effects of shielding. The shields were approximately 2-ft square and were positioned near the plutonium source to effectively shield it from the lab detector. The can of plutonium oxide containing the 108 grams of plutonium was positioned on its side on the floor inside a "cave" of steel and lead bricks. Measurements were then made with the bare source and with the source covered with large slabs of iron and aluminum positioned on top of the can containing the plutonium oxide. The steel and lead cave prevented neutrons from scattering around the edges of the intervening shield materials. Details of the measurements are given in Appendix C, and the results of the shielding measurements are given in Table 3.2. Note that the neutrons were not absorbed like gamma rays in steel shields; the neutrons scattered within the shield and were lower in energy. These data indicate that even 2 in. of steel or aluminum do not greatly influence the measured neutron flux.

3.5 Neutron Measurements in the 324 Building

Operational and background checks were made for neutron levels.

3.5.1 Operational Check

After the preliminary measurements were completed in the laboratory and the system was functioning properly, the holdup measurements commenced in the 324 Building basement in room 3A. Here, the neutron detector was connected to the MCA via 100-foot-long cables, so that the operator could remain in one area while the detector was positioned near the various ducts and piping. Before the

Table 3.2. Effect of Steel and Aluminum Shielding Materials Placed Between the Plutonium and the Slab Neutron Detector

Shield Material	Thickness (in.)	Attenuation Factor
Steel	None	1.00
	1	0.85
	2	0.75
	3	0.68
	4	0.6
	6	0.49
Aluminum	None	1
	0.5	0.94
	1.5	0.84
	2	0.8
	3.5	0.68
	4	0.65

measurements were started, a ^{252}Cf reference source was used as an operational source check of the system. The neutron slab detector was positioned at 12 in. from the source to the front face of the detector. The neutron event spectrum, similar to that shown in Figure 3.2, was measured and recorded every day to verify that the detector and ancillary electronics were functioning properly. After the operational check indicated that the system was still functioning properly, it was ready for neutron measurements on the ducts and pipes.

3.5.2 Background Measurements

Background measurements were performed with an HPGe detector and a neutron counter.

High-Purity Germanium Detector

Background measurements were taken with an HPGe at each measurement location. A solid plug replaced the collimator and a background measurement was taken for the same counting time as the collimated measurement. The background contribution was subtracted from the collimated measurement with analysis conducted on the resulting background-corrected spectrum.

A small, low-power neon laser was used to align the gamma detector to the desired measurement location on the duct, RLWS line, and other measurement locations of interest. Detector-to-source distance was measured using a SONIN sonic measuring unit. Accuracy of the distance measurement was ± 0.5 cm.

Neutron Measurement System

Other neutron sources in the building could contribute significantly to the neutron count rate, so careful background measurements were made before any measurements were made on the ducts or pipes. If the neutron count rates were too high, it would be necessary to modify the measurement plan. The first background measurements were made with the detector in room 3A next to the plutonium storage vault in the basement; the results indicated a background of some 2300 counts in a 5-minute counting period. This background dropped by an order of magnitude in areas far removed from the vault.

The detector was designed to have a directional response, so that fission neutrons entering from the back side would be counted with only half the efficiency of those entering from the front of the slab detector. Background measurements were made on the periphery of the area around the ducts and filter boxes. The slab detector was positioned in a vertical orientation to measure the general background. At a given position, a measurement was taken; then, the slab detector was rotated 180° and the measurement repeated. This procedure gave pairs of neutron count rates with the detector facing the source and pointed away from the suspected neutron source. These data allowed the average room background to be determined, as explained in Section 4.0 on data analysis.

4.0 Analysis of Data

Data was analyzed for both the calibration/verification measurements and for measurements taken in the 324 Building.

4.1 Analysis of Calibration and Verification Measurements

The calibration or verification measurements made before the 324 Building holdup measurements allow one to correlate the mass of weapons-grade plutonium to the measured neutron count rate and account for scatter of neutrons from the walls, floor, and ceiling of the room. This method of correcting for scatter from inverse square measurements is presented in the NBS Special Publication No. 633 (Schwartz and Eisenhauer 1982). The response of any moderated neutron detector can be modeled as a response to neutrons coming directly from the source and a response to neutrons scattered from the floor, walls, and ceiling of the room. If the detector is more than 1 m from the floor and walls, the scattered component can be assumed to be a constant. This can be expressed mathematically by Equation 4.1:

$$C = \frac{M}{r^2} B \quad (4.1)$$

where C = the count rate (cts/s)
 M = the mass of plutonium in grams (directly proportional to the neutron source strength)
 B = the neutron background in the room due to scatter from the floor, walls, and ceiling
 r = the distance (in feet) from the plutonium source to the centerline of the proportional counters.

Multiplying by r^2 gives Equation 4.2:

$$Cr^2 = MBr^2 \quad (4.2)$$

In the limit that r^2 approaches zero, the term Br^2 approaches zero while the term Cr^2 approaches a constant. Plotting the product of the count rate times the distance squared versus the distance squared produces a straight line if the detector is not too close to the source or walls. An example of the inverse square plot from the 108-g plutonium oxide source is shown in Figure 4.1. A straight line has been fitted to the data points. The intercept on the ordinate axis is proportional to the neutron source strength or mass of plutonium, and the difference between the intercept and the value on the straight

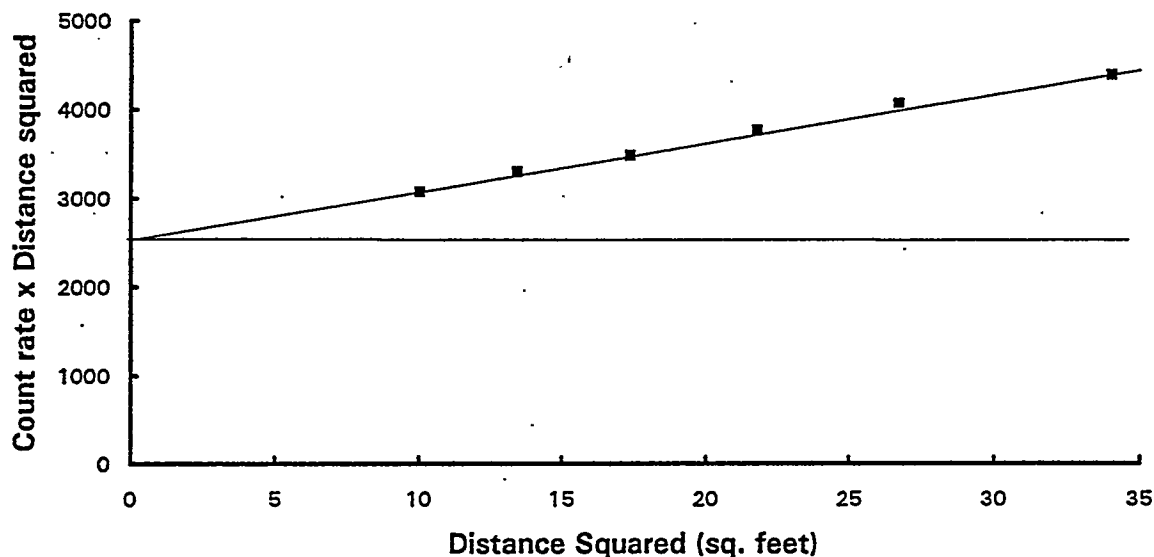


Figure 4.1. Example of Inverse Square Measurements with Neutron Slab Detector Exposed to a Plutonium Oxide Source Containing 108 Grams of Plutonium

line gives the contribution to room scatter. In some small rooms, the scatter can be as high as 50% of the count rate at distances of 6 ft (2 m) from the neutron source. Thus, it is important to correct for the effects of room scatter.

This technique can be used to correlate the detector count rate to the mass of plutonium. Consider the case where the can of plutonium oxide was 4 ft above the floor and the slab neutron detector was positioned vertically. The least squares fit gives the values for the slope and y-intercept of a straight line fitted to the inverse square data points in Figure 4.1. Dividing the slope and intercept values by the mass of plutonium (108 g) gives the results normalized to 1 g. The normalized y-intercept is 0.507, and the normalized slope is 23.66. Putting these values into Equation 4.1 gives an expression for the count rate per gram of plutonium for the slab detector, oriented vertically at 4 ft above the floor. This is expressed mathematically by Equation 4.3:

$$\text{cts/s/g} = 0.507 + \frac{23.7}{r^2} \quad (4.3)$$

where r is the horizontal distance in feet between the plutonium source and the centerline of the ^3He proportional counters at 4 ft above the floor.

4.2 Analysis of 324 Building Measurements

The estimate of the amount of plutonium in the debris at the 324 Building depends on the assumptions made concerning:

- the composition, age, and location or distribution of the plutonium
- the position of shielding materials around the plutonium sources
- the neutron background in the area around the plutonium sources.

4.2.1 Basic Assumptions Used for Calculations

It is possible that there is very little plutonium in the ducts and piping. Many of the measured neutrons could originate from alpha-neutron reactions in low-atomic-number materials, such as borosilicate glass in HEPA filters, and from neutrons from plutonium and curium in the fuel samples in the hot cells that have been scattered into the ducts. It is more probable that the neutrons originate from plutonium and other transuranics (such as ^{241}Am) in the ducts and piping, and great care must be taken in distinguishing between room background and plutonium source neutrons. For all the calculations, the following assumptions were used:

- The neutron emissions were from plutonium and its decay product, ^{241}Am , in the form of an oxide, which is the most stable chemical form. Nitrates, oxylates, hydrated oxides, and hydroxides would produce similar neutron yields. Neutrons from other transuranics were ignored. This results in conservative estimates for the mass of plutonium.
- There is little interference from other plutonium compounds, such as fluorides, or from alpha-neutron reactions with low-atomic-number elements, such as beryllium, boron, aluminum, or sodium. It is difficult to judge whether there is a significant contribution from alpha-neutron reactions without more sophisticated measuring equipment that can measure two neutrons in coincidence from a single fission event. (Alpha-neutron events would produce only random coincidences.)
- Each plutonium source is treated separately and does not interact with other plutonium sources (i.e., there is no multiplication).
- Any multiplication effects were ignored, which is a good assumption for gram quantities of plutonium. There may have been some increase in the neutron emissions from the 108-g plutonium sample used as the verification standard, but any errors introduced will be small in comparison with other sources of error or uncertainty.
- Any plutonium holdup is low-exposure plutonium with very little ^{240}Pu . The weapons-grade plutonium from LLNL (ash sample RA 146A) is assumed to be representative of the plutonium in the debris. This is a reasonable assumption and will produce conservative estimates. Any plutonium present with a higher ^{240}Pu content will emit more neutrons per gram; consequently, the mass of plutonium will be overestimated.

4.2.2 Neutron Background

Before the measurements on the ducts and pipes were initiated, some measurements were made to determine the neutron backgrounds of the periphery of the area. When a source was located, two measurements were made: one measurement with the front face of the detector pointing towards the suspected plutonium source, and a second measurement with the slab detector pointing away from the source. Measurements were made with the slab neutron detector oriented at right angles to the suspected source, i.e., the long axis of the slab was perpendicular to the source. In general, the effective "field of view" was about 3 to 4 ft at a distance of 1 to 2 ft from the suspected plutonium source. The following discussion demonstrates how it is possible to estimate the effects of room background with the slab detector oriented at right angles to a suspected plutonium source in the ducts or piping.

Consider the example of the bottom prefilter box on the west side of the SERF cell in the 327 Building basement. The slab neutron detector was positioned directly to the side of the prefilter box at a distance of 12 in. from the surface of the iron shield placed around the box. With the front face of the slab detector pointed towards the prefilter box, the measured neutron count rate was 8.84 ± 0.27 cts/s. When the detector was rotated 180° so that the front of the detector was pointed away from the filter, the measured count rate was 4.48 ± 0.19 cts/s. We can be reasonably certain that there were no neutron sources directly in front of the slab detector when it was pointed away from the filter box.

Fission neutrons entering from the back of the detector were counted with about 53% of the efficiency of neutrons entering from the front face, as shown by the experimental data for angular efficiency discussed in Section 3.4.3. Let x represent the counts from the source and y represent the counts from room background. The count rate measured with the vertical slab detector pointed toward the suspected source is the sum of the counts from neutrons entering the front and 50% of the neutrons entering from the back:

$$x + 0.50y = 8.84 \text{ cts/s} \quad (4.4)$$

Likewise, the count rate measured with the vertical detector pointing away from the prefilter box and facing the room is the sum of the neutrons entering from the front (or room background neutrons) and 50% of the neutrons entering from the back side:

$$y + 0.50x = 4.48 \text{ cts/s} \quad (4.5)$$

Solving the two simultaneous equations for x and y gives:

$$\begin{aligned} y &= 0.08 \text{ cts/s for neutrons from room background} \\ x &= 8.80 \text{ cts/s for neutrons emanating from the filter box.} \end{aligned}$$

Thus, we must correct the measured count rates by subtracting away 0.08 cts/s due to the neutron background in the room. This is a selected example in which the plutonium in the filter box is well isolated and the count rates are much higher than the general room background. In every other

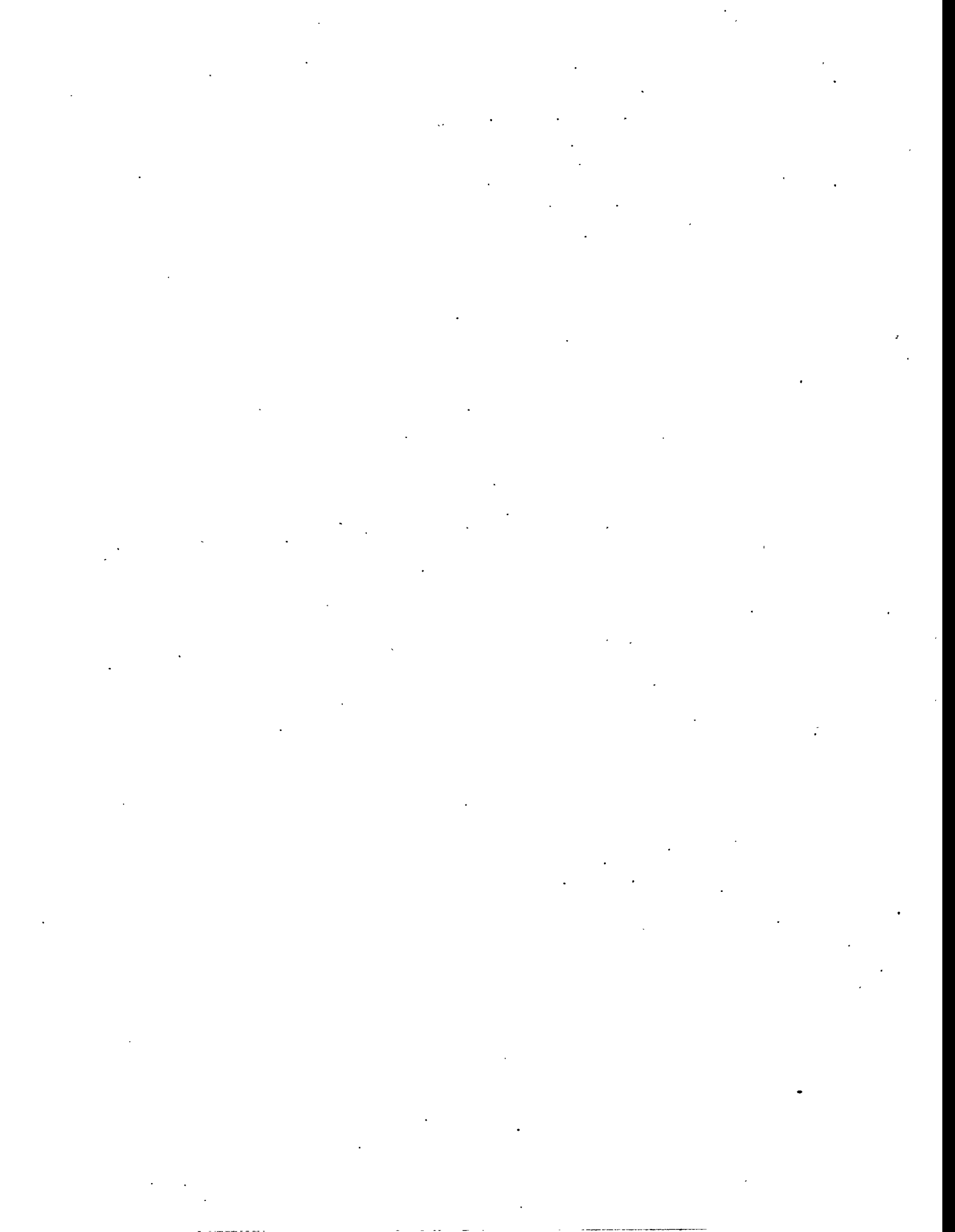
measured location, the count rate with the slab detector pointed towards the suspected plutonium source is similar to the count rate with the detector pointed away from the source. The background-corrected count rates are given in the data table in the spreadsheet calculations in Appendix C.

4.2.3 Calculated Mass of Plutonium

Once the contribution from room background is calculated (or estimated from measurement in the general area), it is possible to use Equation 4.3 to estimate the effects of room scatter and determine the mass of weapons-grade plutonium (6% ^{240}Pu) in the source, if the distance to the center of the plutonium source is known. In the case of the prefilter box, discussed in the previous section, the dimensions of the box were 31.5 in. long by 31 in. high: the box was shielded by 2.5 in. of iron. The center of the slab neutron detector was 12 in. from the surface of the iron shield covering the filter box. If we assume that the plutonium is uniformly spread over the filter, the plutonium source can be modeled as a point source at a distance of $31.5/2 + 2.5 + 12 = 30.25$ in. (2.52 ft) from the center of the detector. Substituting a distance of 2.52 ft into Equation 4.3 gives:

$$0.507 + \frac{23.7}{(2.52)^2} = 4.24 \text{ cts/s/g} \quad (4.6)$$

Dividing the measured count rate of 8.80 cts/s by 4.24 cts/s/g of plutonium gives a value of 2.1 g of plutonium in the filter box. It may be somewhat suspect because alpha-neutron reactions from finely divided alpha-emitters on borosilicate glass fiber can produce elevated neutron levels. Because the neutron emission rates from higher-exposure plutonium are higher than for weapons-grade plutonium, the value of 2.1 g of plutonium is an upper limit on the amount of plutonium that may be in the prefilter box.



5.0 Results

The results of this survey indicate no significant levels of plutonium reside in the ductwork or RLWS lines in the 324 Building. Appendix C contains the results of neutron measurements; Appendix D shows the gamma measurements.

5.1 Neutron Measurement Results

Neutron measurements of the 324 Building ductwork and RLWS lines were taken at equal distances along the suspected source using the neutron slab detector. Corrections for overlapping fields were applied to the data. Two sets of plutonium mass estimates were determined using an assumed weapons-grade isotopic composition for the maximum plutonium mass and a reactor fuel isotopic composition (11% ^{240}Pu) for the most likely mass. Table 5.1 details the results of the neutron measurements by room. Table 5.2 lists the neutron measurement results specifically for the RLWS lines in the service tunnel. Special attention was given to "hot spots" with high gamma activity, which were thought to be fuel particles in the RLWS pipe.

Table 5.1. Plutonium Holdup in Ductwork and RLWS Lines

Location	Maximum Plutonium (g) ^(a)	Most Likely Plutonium (g) ^(a)
Room 3A	10.71	5.85
Room 3B	1.34	0.73
Room 3C	0.20	0.11
Room 3F	1.25	0.68
Room 3J	0.26	0.14
Room 3K	1.05	0.57
Room 4/4A	0.27	0.15
Room 146	1.28	0.70
Room 147	2.21	1.21
Tank Pit	0.47	0.26
Total	19.04	10.40

(a) Maximum and most likely plutonium mass estimates are based on 6 and 11 weight percent ^{240}Pu , respectively.

Table 5.2. Plutonium Holdup in Radioactive Liquid Waste System Pipeline in Service Tunnel

<u>Location</u>	<u>Maximum Plutonium (g)^(a)</u>	<u>Most Likely Plutonium (g)^(a)</u>
ST-3	0.02	0.01
ST-6	0.02	0.01
ST-9	0.03	0.02
ST-12	0.02	0.01
ST-13	0.02	0.01
ST-17	0.01	0.01
ST-20	0.04	0.02
ST-34	0.06	0.03
ST-39	0.05	0.03
ST-43	0.02	0.01
ST-44	0.02	0.01
ST-48	0.01	0.01
ST-50	0.01	0.01
ST-51	0.01	0.01
ST-53	0.03	0.02
Total in RLWS Line	0.37	0.20

(a) Maximum and most likely plutonium mass estimates are based on 6 and 11 weight percent ²⁴⁰Pu, respectively.

Totals for all neutron measurement locations are
 maximum 19.41 g
 most likely 10.6 g.

Other neutron measurement results not included in the overall total are listed in Table 5.3. These measurements were taken in Room 18 directly under the hot cell operating gallery, and are suspect

Table 5.3. Estimation of Plutonium in the Service Gallery (Room 18) by Neutron Measurement

<u>Location</u>	<u>Maximum Plutonium (g)^(a)</u>	<u>Most Likely Plutonium (g)^(a)</u>	<u>Description of Location</u>
18-1,-3	8.57	4.68	Contaminated area next to cell wall (old leak)
18-2	79.21	43.25	Penetrations to duct space
18-4,-8	0.30	0.16	Floor drains
18-5,-7	0.40	0.22	RLWS line
18-9	0.07	0.04	Crib waste stub
Totals	88.55	48.35	

(a) Maximum and most likely plutonium are based on 6 and 11 weight percent ²⁴⁰Pu, respectively.

because of the high neutron background from the hot cells and adjacent duct space containing the HEPA filter banks for the hot cells. Since these neutrons are most likely coming from fissionable material in areas that are not accessible to personnel and therefore are outside the scope of this study, these quantities are not included in the totals for the 324 building.

5.2 Gamma Measurement Results

A summary of ^{137}Cs curie content in the various ducts and pipes is presented in Table 5.4. These data show the survey instrument result and estimated activity level at each location. Appendix B contains figures that show the position of the measurements and Appendix D contains the spreadsheets with the measurement results. The curie contents of the RLWS lines in the service tunnel are shown separately in Table 5.5.

Table 5.4. ^{137}Cs Curie Content By Direct Gamma Ray Measurement Using a Collimated High-Purity Germanium Detector

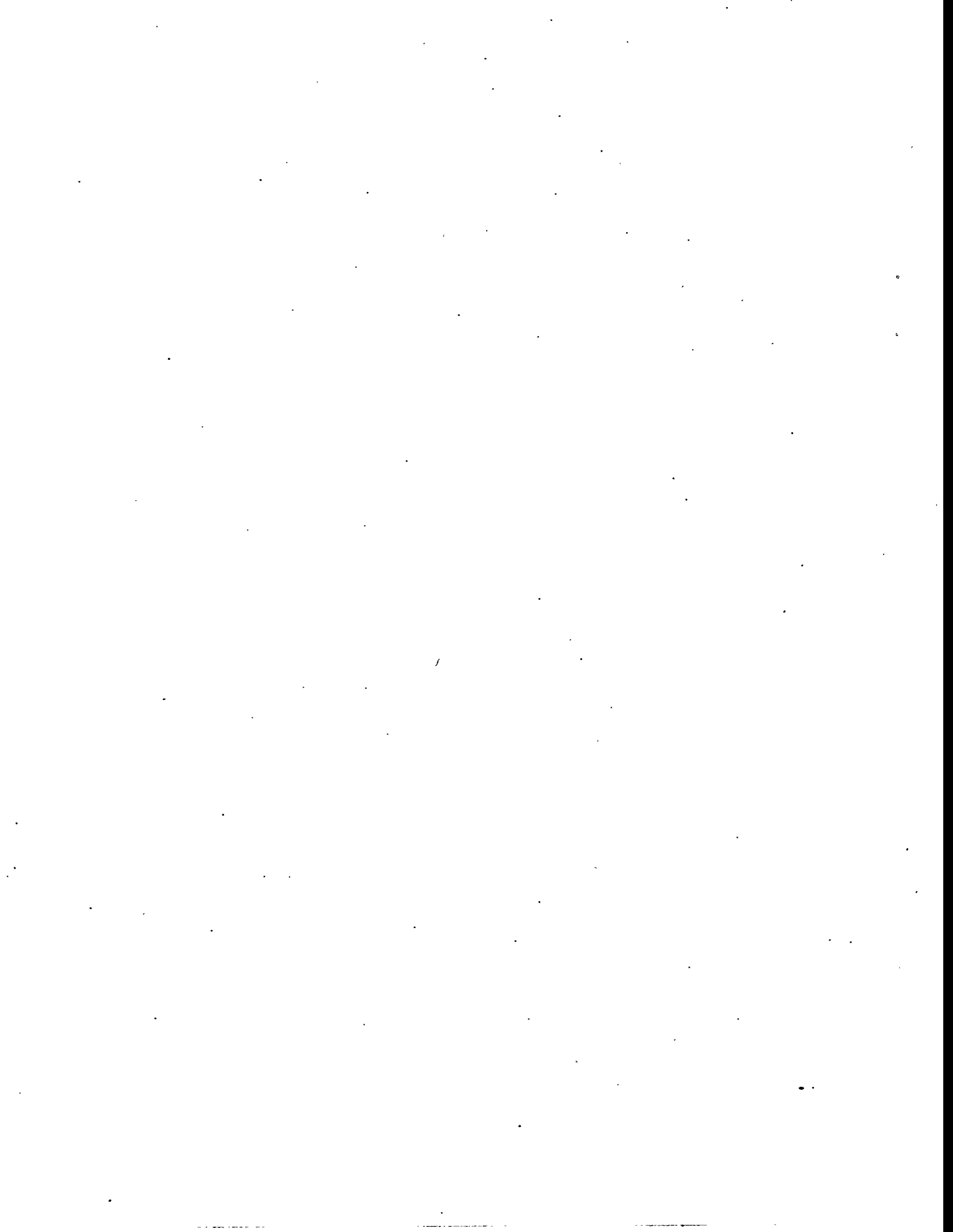
<u>Location</u>	<u>Distance (in.)</u>	<u>Count Rate (cpm)</u>	<u>$\mu\text{Ci } ^{137}\text{Cs}$</u>
3A-1	33	7.20	0.26
3A-2	36	72.00	3.13
3A-3	36	28.4	1.24
3A-Tank	36	29.80	1.30
3K-1&2	36	182.94	7.96
3K-4&5	36	51.48	2.24
4-17	26	326.76	7.42
4-18	36	1194.18	51.99
4-19	43	47.40	2.94
4-Tank	36	289.56	<u>12.61</u>
		Total	91.09

Table 5.5. Radioactive Liquid Waste System Pipeline Activity Profile

RLWS Line 1					
Location	Distance	Count Rate (cpm)		¹³⁷ Cs	μCi/section
		Top	Bottom		
1	3	1800	6500		48.3
2	5.5	900	2400		21.4
3	8.5	400	1700		10.5
4	11	1000	3000	5.86	17.7
5	13	500	2000		10.4
6	15	1000	3500		40.2
7	19	400	1800		8.9
8	21	600	2000	2.08	10.8
9	23	1000	3000		35.4
10	27	400	1000		10.0
11	31	400	1000		7.5
12	34	1000	3000		13.3
13	35.5	1000	3500		20.1
14	37.5	400	1000		10.0
15	41.5	400	1000		5.0
16	43.5	400	1000		6.2
17	46	1000	4000	2.39	22.6
18	48	400	1000		5.0
19	50	400	1000		10.0
20	54	400	1000	2.79	10.0
21	58	400	1000		5.0
22	60	400	800		4.0
23	62	400	600	1.38	3.0
24	64	400	1000		10.0
25	68	400	1000		10.0
26	72	400	1000		5.0
27	74	400	1800		17.8
28	78	400	1000		7.5
29	81	1000	3000		35.4
30	85	400	1000		10.0
31	89	400	1000		10.0
32	93	400	1000		10.0
33	97	400	1000		10.0
34	101	400	1000		10.0
35	105	400	1000		10.0
36	109	400	800		8.0
37	113	400	800		8.0
38	117	400	800		10.0

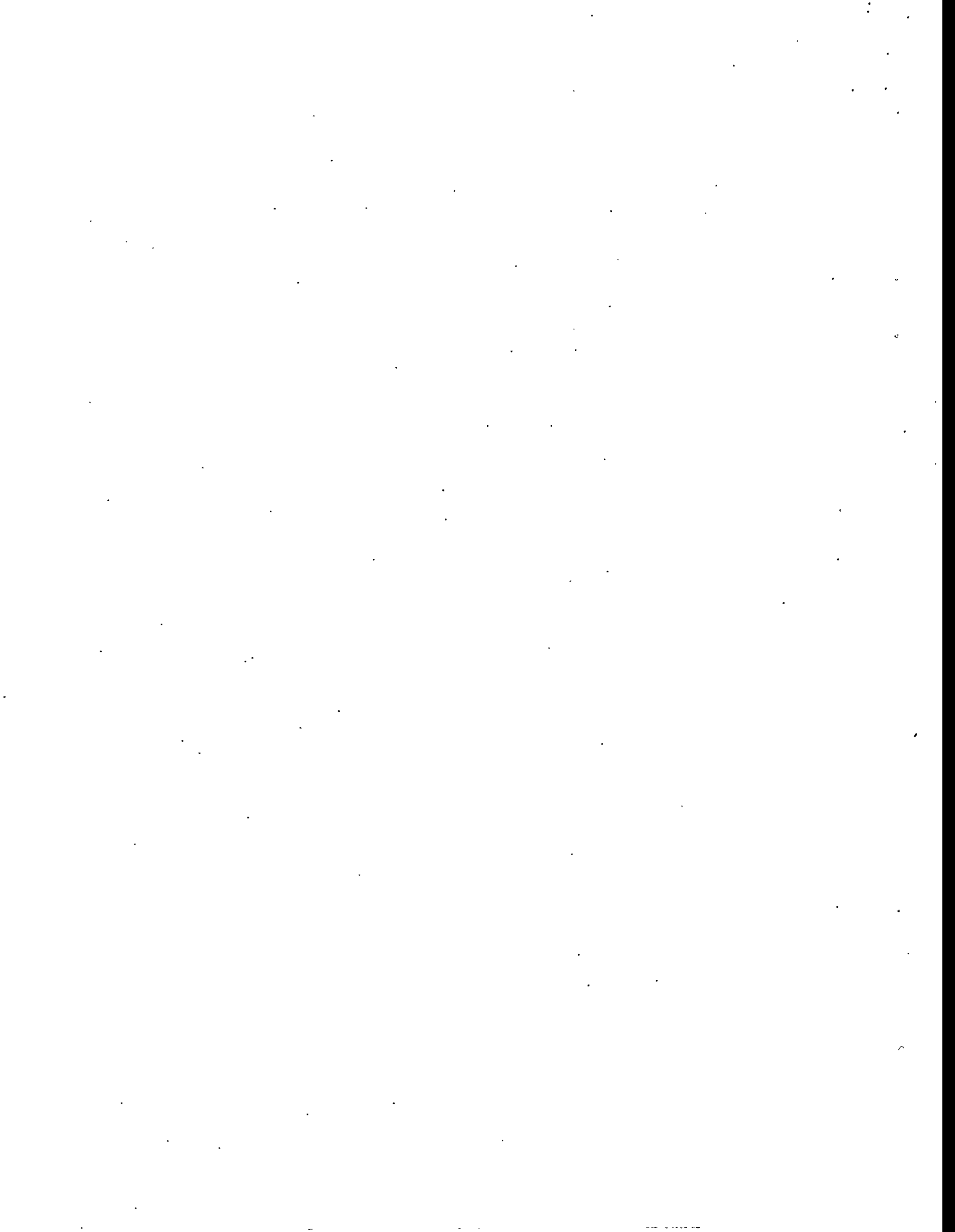
Table 5.5. (contd)

RLWS Line 1					
Location	Distance	Count Rate (cpm)		¹³⁷ Cs	μCi/section
		Top	Bottom		
39	122	1000	1800		23.6
40	126	400	800		8.0
41	130	400	800		8.0
42	134	400	800		6.0
43	137	400	800		6.0
44	140	400	800	29.31	4.0
	142				
Subtotal in RLWS 1					562.0 μCi
RLWS Line 2					
Location	Distance	Count Rate (cpm)		Cs-137	μCi/section
		Top	Bottom		
45	10	200	200	0.8	
46	26	200	200	0.14	1.0
47	46	200	200	0.54	0.5
48	56	200	200		0.2
49	60	200	400		3.8
50	67	2000	6000	125.42	250.2
		0			
51	71	1000	4000	71.84	232.8
		0			
52	78	300	200	1.75	1.2
53	82	1200	3500	19.87	21.1
54	84	100	300		0.9
55	103	375	500		8.5
56	110	100	300		0.2
57	114	200	400		5.4
58	124	200	400		10.7
59	144	200	300		0.6
	146				
Subtotal in RLWS 2					537.8 μCi
Total in RLWS 1 + 2					1099.8 μCi



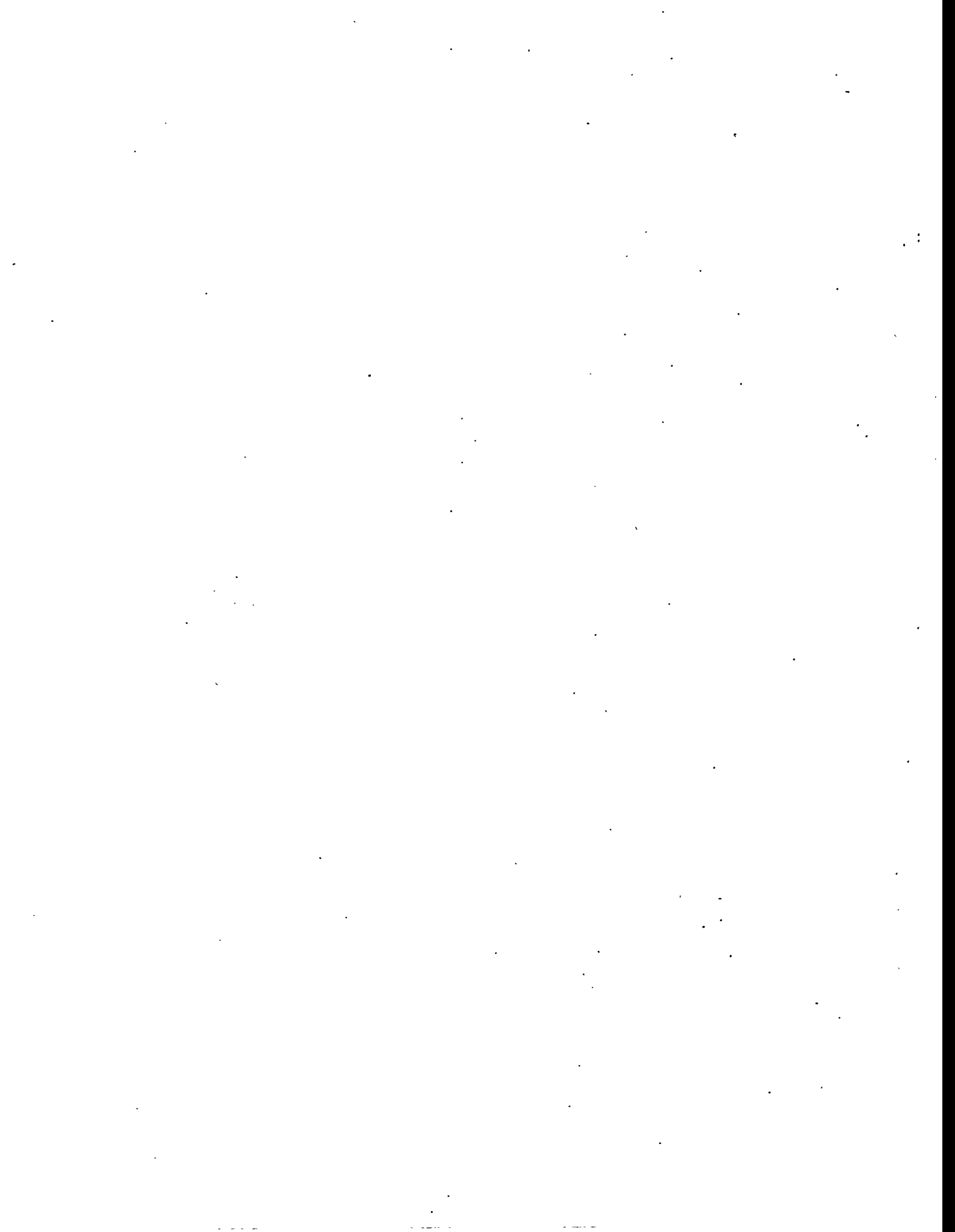
6.0 References

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- U.S. Nuclear Regulatory Commission (NRC). 1984. *In Situ Assay of Plutonium Residual Holdup*. U.S. Nuclear Regulatory Guide 5.23, U.S. Nuclear Regulatory Commission, Washington, D.C.



Appendix A

Basic Holdup Assay Measurements Procedure MCA-510



Appendix A

Basic Holdup Assay Measurements Procedure MCA-510

1. The assay system components are set up in the laboratory and pretest verifications are performed.
2. Perform energy calibration and measurement control procedure, completing Exhibit A (see Figure A.1).
3. Perform calibrations for the assay scenario.
4. Move instrumentation to location and verify that no damage has occurred. Perform an energy calibration and measurement control procedure, completing Exhibit A.
5. Determine ambient background(s) at the counting location and evaluate counting technique(s).
6. Perform measurements with intermittent control checks, record data and location, and assign non-destructive assay (NDA) log numbers.
7. Transfer data and analyze, using software acquisition and analyses codes and computer spreadsheets. Retain the original data for archive, logging the file name on the Exhibit B, Pacific Northwest Laboratory (PNL) NDA Analysis Log/Report (see Figure A.2).
8. Evaluate the data results and determine the appropriate correction factors, including the detector filters. Initial basic assumptions are made of the source material, one being that the sample self-attenuation is negligible. Actual attenuation correction factors can be applied after measurements are made and more reasonable assumptions are determined as to the material matrix and configuration. All counts measured are corrected by the attenuation correction factors (CF).

Generally, a density value in grams per cubic centimeter (g/cm^3) is derived for the item based on gross weight of the container minus the container weight and/or shielding and/or packaging materials. This matrix density value is used for the attenuation corrections for the gamma energies measured.

9. Perform the analysis, review the results, and issue a report.

PNL NONDESTRUCTIVE ASSAY MEASUREMENT PROCEDURE

Procedure No.: NDA-510 Revision: 1 Effective Date: July 26, 1994 Page 31 of 34

EXHIBIT A

M & TE SET-UP AND MCA ENERGY CALIBRATION CHECKLIST

Date _____ Location _____ Work Pkg # _____ Operator _____ Time _____

List Assayed Items(s) and NDA Log #:

SET-UP CHECKLIST

- | | | |
|-----|---|----|
| (1) | Review RWP, CSP | () |
| (2) | Gamma Detector Filled With LN, Stabilized | () |
| (3) | Filters on Detector Y / N List _____ | () |
| (4) | Gamma Detector # _____ | () |
| (5) | HV Supply Settings; Pos _____ Neg _____ | |
| (6) | Neutron Detector Mod/Ser. _____ | |
| (7) | HV Supply Settings; Pos _____ Neg _____ | |

ENERGY CALIBRATION

(7) Spectroscopy Amplifier Settings:

Model No. _____	
Course Gain _____	30
Threshold _____	AUTO
Polarity Input _____	NEG
Polarity Output _____	POS
Shaping Time _____	2 μ sec
Fine Gain _____	
Shaping Multiplier _____	1

(10) ADC Settings

Model/Serial No. _____	
Gain _____	4096
Range _____	4096
Digital Off Set _____	NONE
Coincidence Gate II _____	NONE

(11) MCA Identification

Model/Serial No. _____

(12) Neutron System

Model/Serial No. _____	
Counter Model/Serial No. _____	
Threshold Setting _____	

(13) Gamma Measurement Control

Initial PreTest	Count Time _____
Peak Energy _____	Net Area _____
_____	_____
_____	_____
Final Post Test	Count Time _____
Peak Energy _____	Net Area _____
_____	_____
_____	_____

(9) Systems Adjustments and Noise

With Oscilloscope at 10 μ sec/div ()

Check Pole Zero ()
 Check Peak Shape ()

SPECIALIST REVIEW/DATE _____

Figure A.1. Calibration Checklist

PNL NONDESTRUCTIVE ASSAY MEASUREMENT PROCEDURE

Procedure No.: NDA-510 Revision: 1 Effective Date: July 26, 1994 Page 32 of 34

EXHIBIT B

PNL NDA ANALYSIS ASSAY LOG/REPORT

DATE: _____ NDA LOG # _____
TRU U MFP Activation SAMPLE # _____
WEIGHT #/G _____

MEASUREMENT SYSTEMS/METHODS:

- ___ Gamma Energy Analysis GEA
- ___ Calorimeter
- ___ Gamma Isotopics
- ___ Passive Neutron Coincidence Well Counting
- ___ Active Neutron Well Counting
- ___ Passive Neutron Counting / Spectrometry
- ___ Segmented Gamma Assay (Transmission Corrected)
- ___ Combined Neutron Coincidence/Segmented Gamma Assay (Transmission Corrected)
- ___ Far Field Assay Techniques
 - ___ Barrels
 - ___ Process Hold-up, HEPA Filters
 - ___ Waste Boxes
 - ___ Project Specific
 - ___ Other _____

Measurement Information	
	Dose Rate _____

ASSAY RESULTS: Report attached Y / N Issue Date _____

ISOVER Report Y / N Issue Date _____

DATA ARCHIVED: Disk(s) Directory / Prefix _____

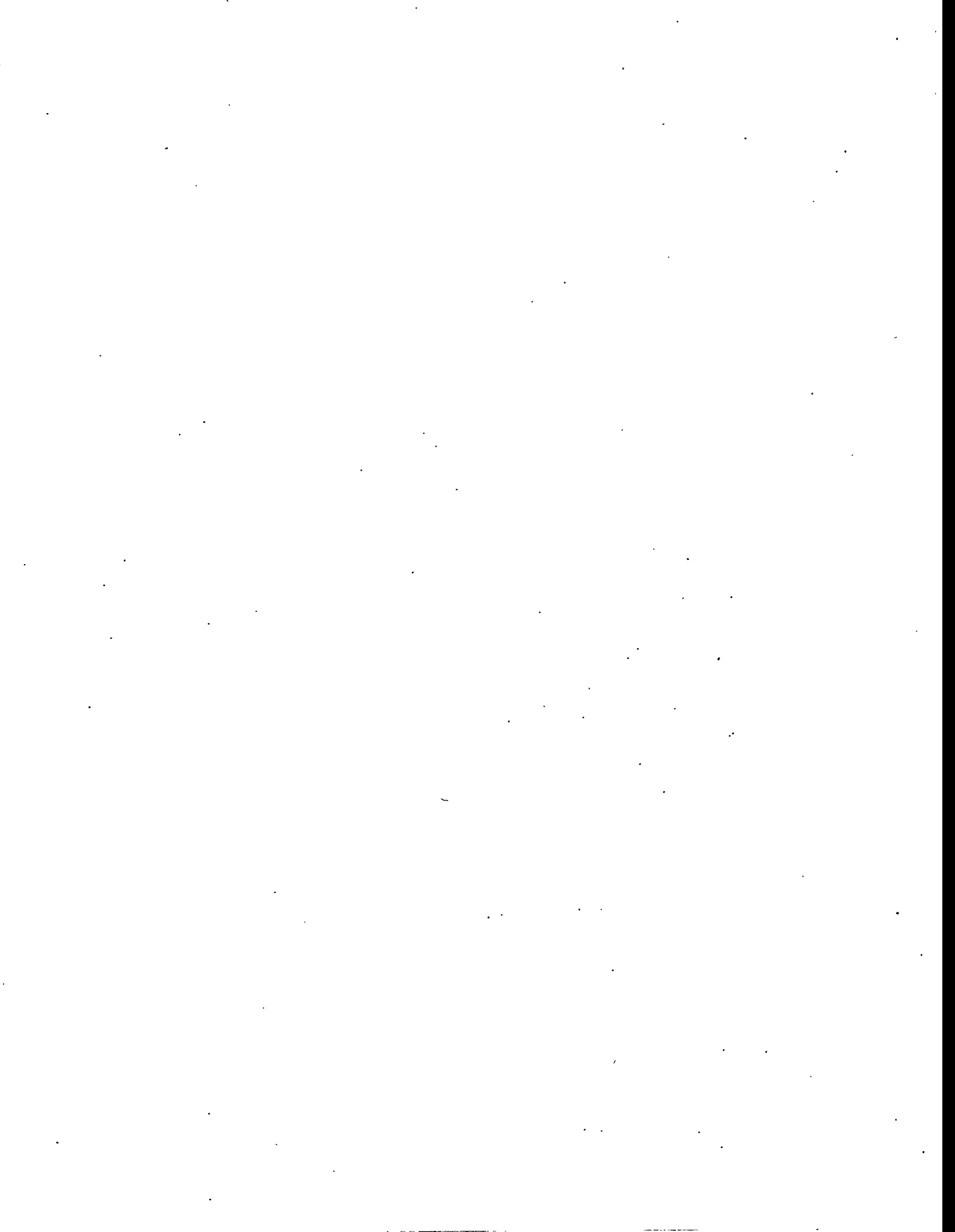
File Names _____

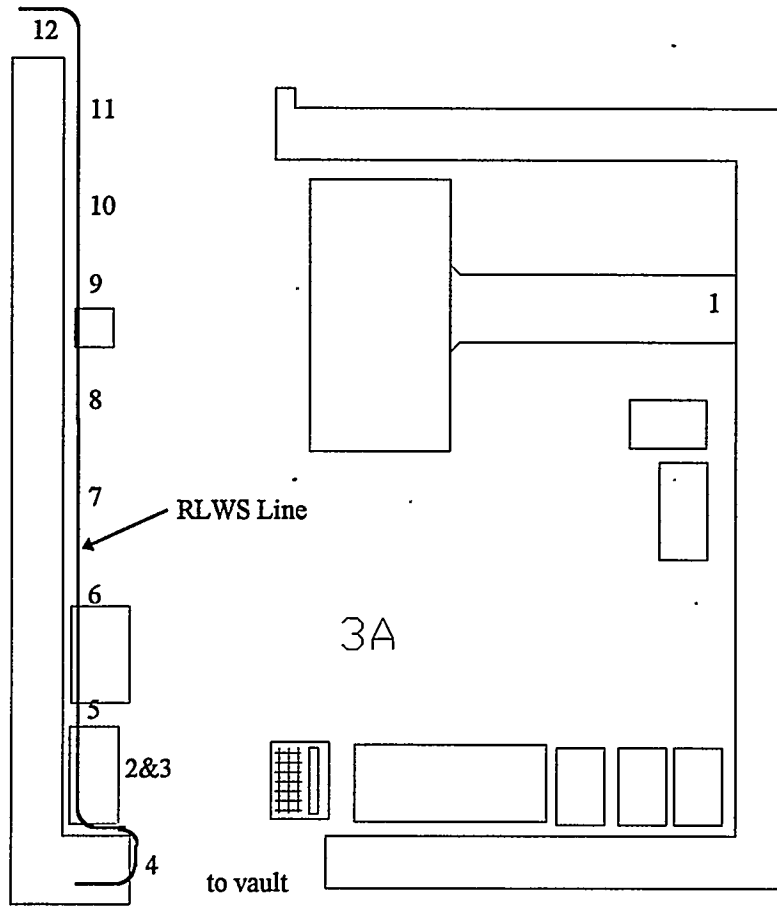
Specialist Review _____

Figure A.2. Assay Log/Report Form

Appendix B

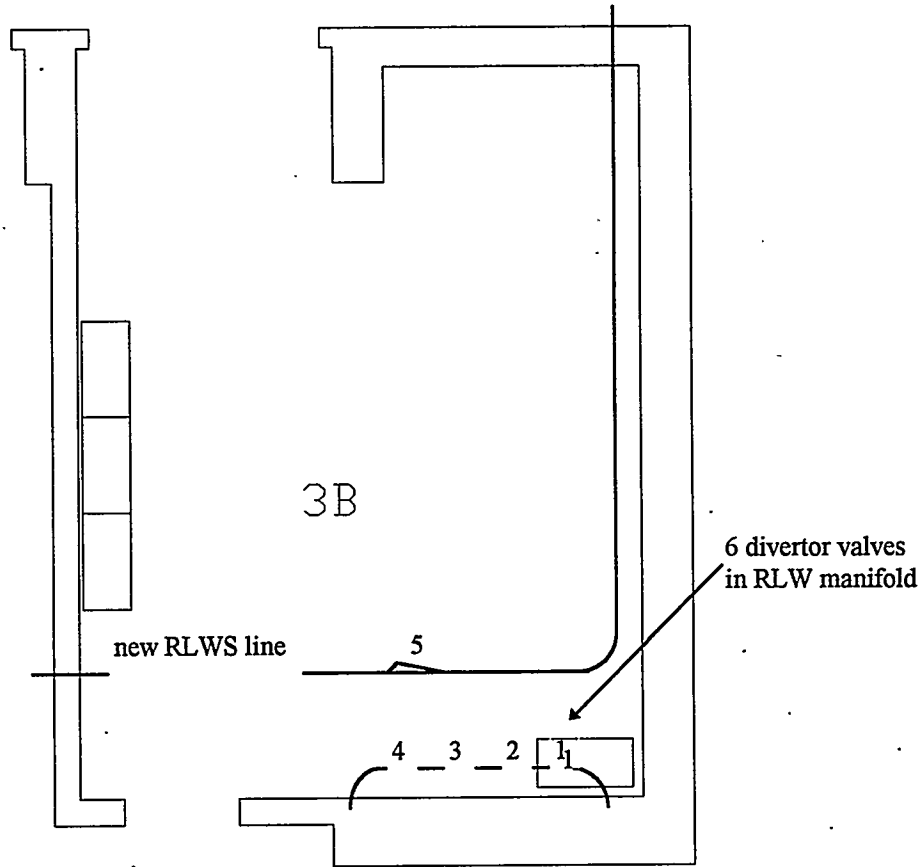
Data Sheets for Radiological Survey





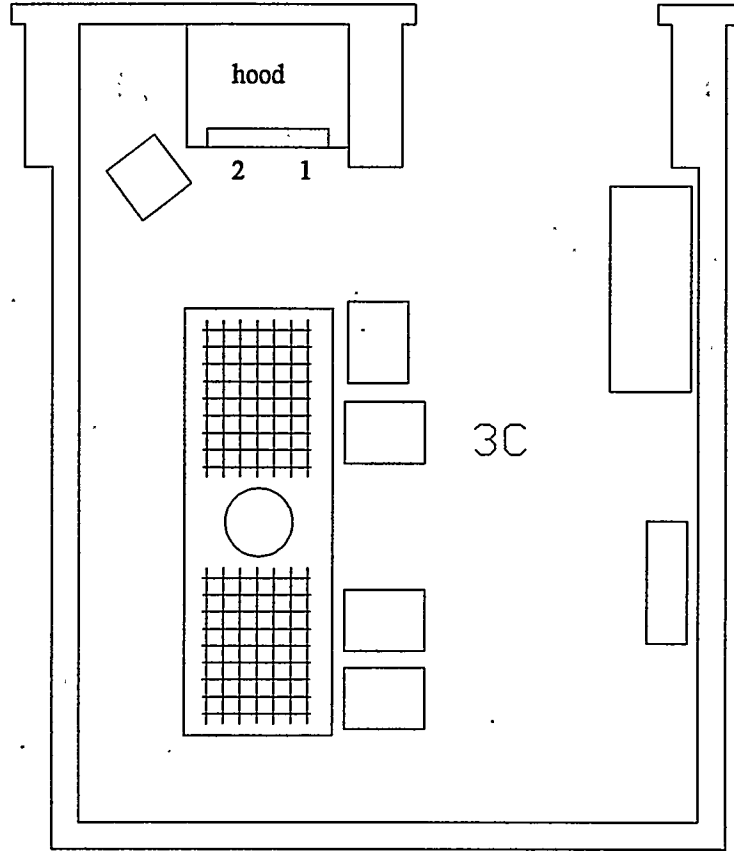
Measurement Locations

Locations	Top	Bottom	Comments
3A_1	<u>150 c/m</u>	<u>150 c/m</u>	<u>old duct flange at wall</u>
3A_2	_____	<u>400</u>	<u>2' above floor</u>
3A_3	_____	<u>200</u>	<u>4' above floor</u>
3A_4	<u>2000</u>	<u>2000</u>	<u>RLWS line on floor</u>
3A_5	_____	<u>200</u>	<u>RLWS line - valve</u>
3A_6	_____	<u>400</u>	<u>RLWS line - behind camera</u>
3A_7	_____	<u>400</u>	<u>RLWS line</u>
3A_8	_____	<u>500</u>	<u>RLWS line</u>
3A_9	_____	<u>500</u>	<u>RLWS line</u>
3A_10	_____	<u>400</u>	<u>RLWS line</u>
3A_11	_____	<u>350</u>	<u>RLWS line - by door</u>
3A_12	<u>37,000</u>	_____	<u>Really in room 3J</u>



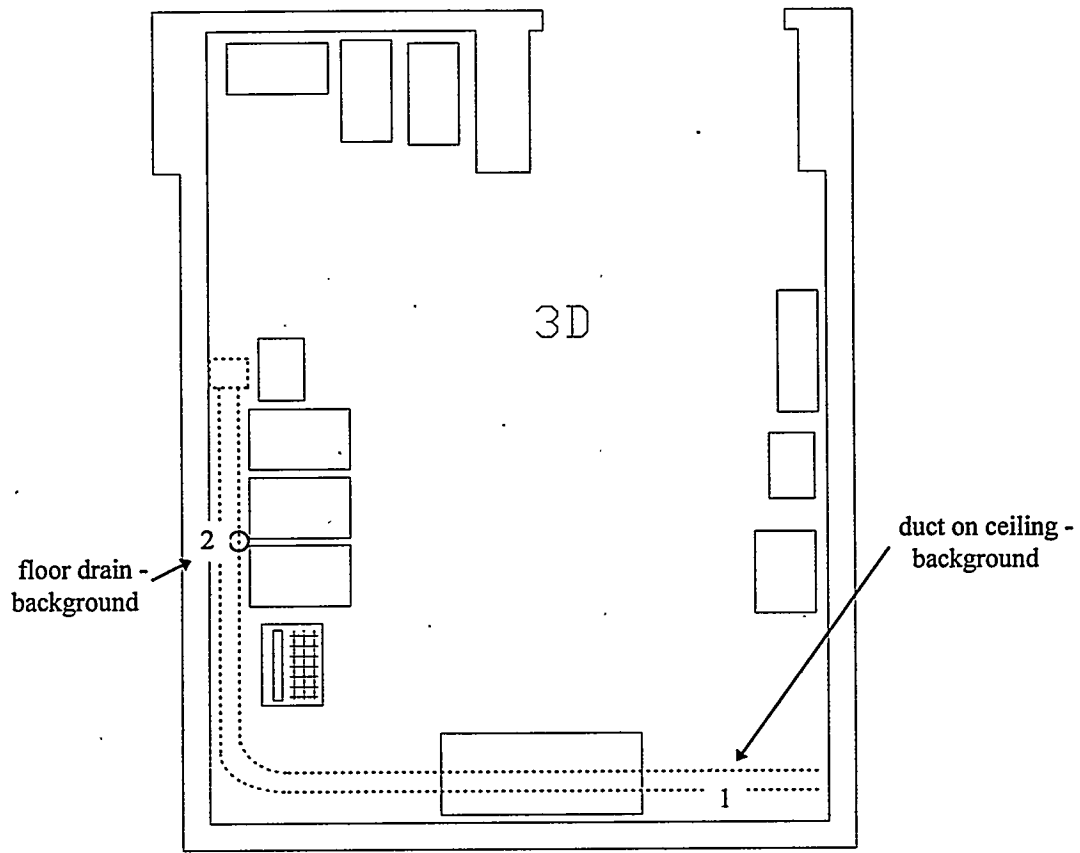
Measurement Locations

Locations	Top	Bottom	Comments
3B_1	<u>10.000</u>	<u>20.000</u>	<u>divertor valves</u>
3B_2	<u>10.000</u>	<u>20.000</u>	<u>divertor valves</u>
3B_3	<u>10.000</u>	<u>20.000</u>	<u>divertor valves</u>
3B_4	<u>10.000</u>	<u>20.000</u>	<u>divertor valves</u>
3B_5	<u>bkgd</u>	<u>bkgd</u>	<u>largest neutron repsonse</u>



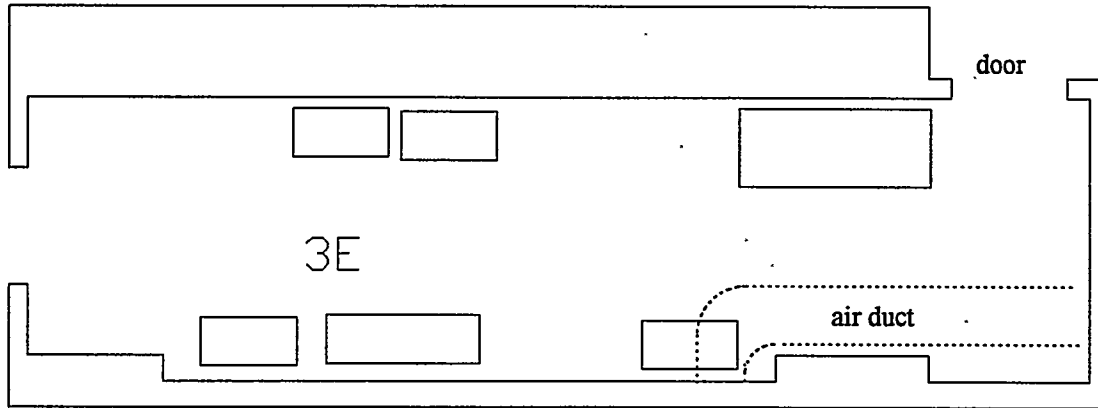
Measurement Locations

Locations	Top	Bottom	Comments
3C_1	<u>100</u>	<u>100</u>	<u>Fume hood</u>
3C_2	<u>100</u>	<u>100</u>	<u>Fume hood</u>



Measurement Locations

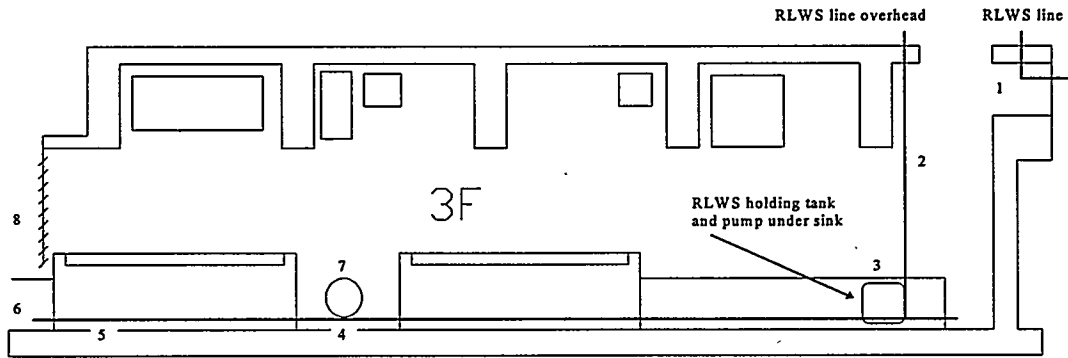
Locations	Top	Bottom
3D_1	_____	<u>bkgd</u>
3D_2	_____	<u>bkgd</u>



nothing above
background
in air duct

Measurement Locations

Locations	Top	Bottom
3E_1	_____	_____
3E_2	_____	_____
3E_3	_____	_____



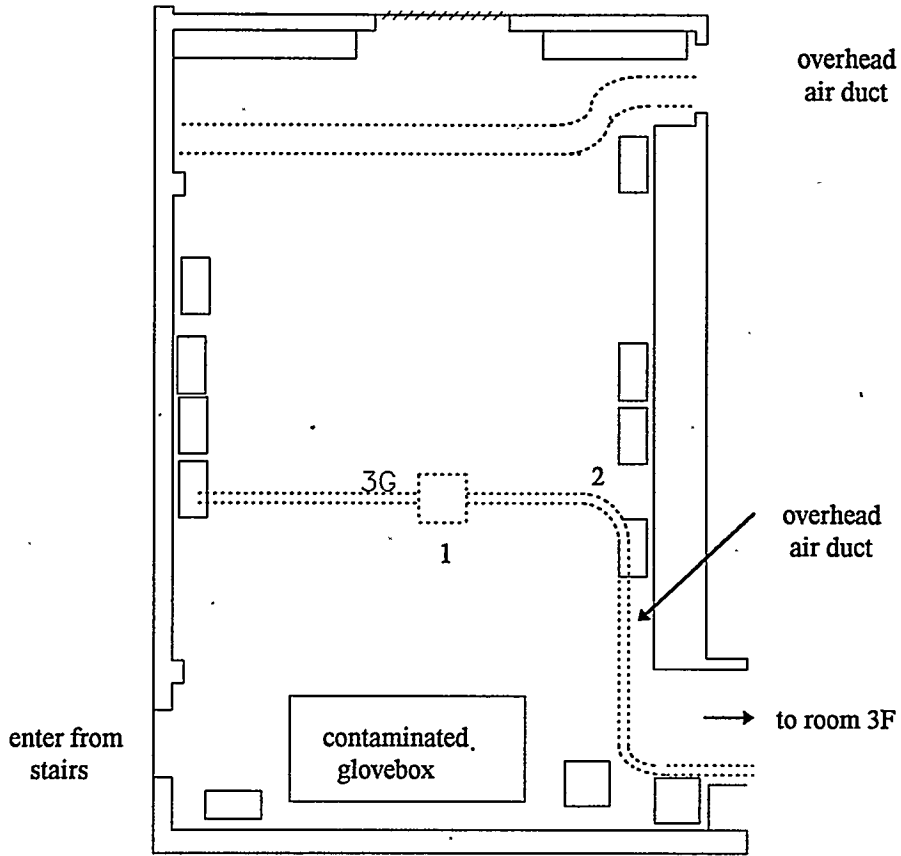
Measurement Locations

Locations	Top	Bottom	Comments
3F_1	<u>70,000 c/m</u>	<u>70,000 c/m</u>	<u>elbow of RLWS line penetration in wall</u>
3F_2	<u><200 c/m</u>	<u><200 c/m</u>	<u>RLWS line, 4' from elbow in pipe</u>
3F_3	<u><200 c/m</u>	<u><200 c/m</u>	<u>RLWS holding tank - 1' from tank</u>
3F_4	<u><200 c/m</u>	<u><200 c/m</u>	<u>RLWS line in space between hoods</u>
3F_5	<u>1100 c/m</u>	<u>hot spot</u>	<u>RLWS line 11.5" from wall</u>
3F_6	<u><200 c/m</u>	<u><200 c/m</u>	<u>12' to tee in RLWS line</u>
3F_7	<u>too high</u>	<u>to reach</u>	<u>overhead air duct between hoods</u>
3F_8	<u>too high</u>	<u>to reach</u>	<u>Tee of all air ducts from hoods & room</u>

Data Entered by: _____

Date: _____

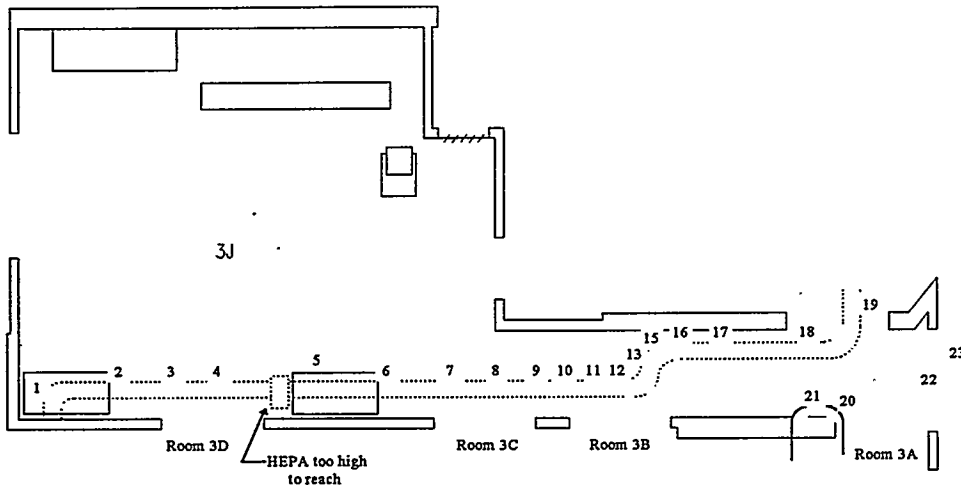
Con



Measurement Locations

Locations	Top	Bottom	Comments
3G_1	<u>background</u>	_____	<u>old inlet for return air</u>
3G_2	<u>background</u>	_____	<u>elbow - 110" above floor</u>

Note: Same Eberline E140B used for rooms 3E, 3F, and 3G

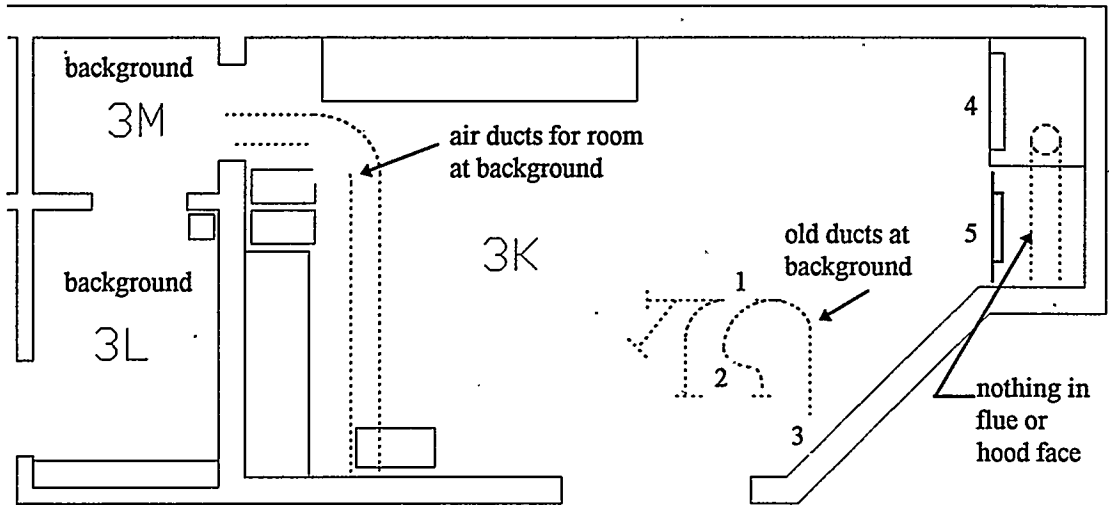


Measurement Locations

Locations	Top	Bottom	Comments
3J_1		<u>4000</u>	<u>air duct from SMF</u>
3J_2		<u>900</u>	<u>air duct</u>
3J_3		<u>700</u>	<u>air duct</u>
3J_4		<u>700</u>	<u>air duct</u>
3J_5		<u>1000</u>	<u>air duct</u>
3J_6		<u>800</u>	<u>air duct</u>
3J_7		<u>700</u>	<u>air duct</u>
3J_8		<u>900</u>	<u>air duct</u>
3J_9		<u>900</u>	<u>air duct</u>
3J_10		<u>1000</u>	<u>air duct</u>
3J_11		<u>1700</u>	<u>air duct</u>
3J_12		<u>1800</u>	<u>air duct</u>
3J_13		<u>2100</u>	<u>air duct</u>
3J_14		<u>600</u>	<u>air duct</u>
3J_15		<u>350</u>	<u>air duct</u>
3J_16		<u>350</u>	<u>air duct</u>
3J_17		<u>350</u>	<u>air duct</u>
3J_18		<u>200</u>	<u>air duct</u>
3J_19		<u>250</u>	<u>air duct</u>
3J_20		<u>37,000</u>	RLWS line - weld in pipe 26" to wall
3J_21		<u>700</u>	RLWS line - middle of pipe
3J_22		<u>300</u>	RLWS line - maze of pipes by 4A door
3J_23		<u>400</u>	air duct at door of 4A

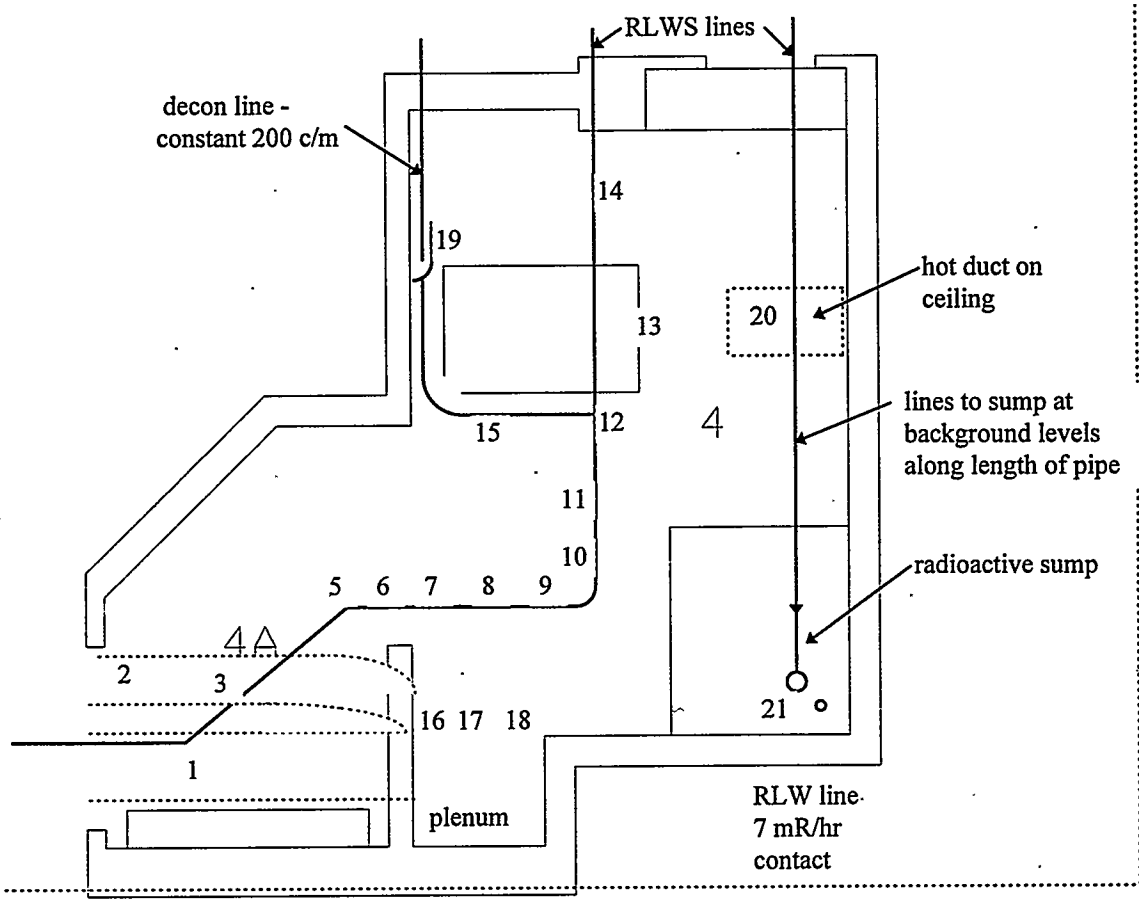
Data Entered by: _____

Date: _____



Measurement Locations

Locations	Top	Bottom	Comments
3K_1	_____	<u>100</u>	<u>old glove box duct</u>
3K_2	_____	<u>background</u>	<u>detector centered between pipes</u>
3K_3	_____	<u>background</u>	
3K_4	_____	<u>background</u>	
3K_5	_____	<u>background</u>	

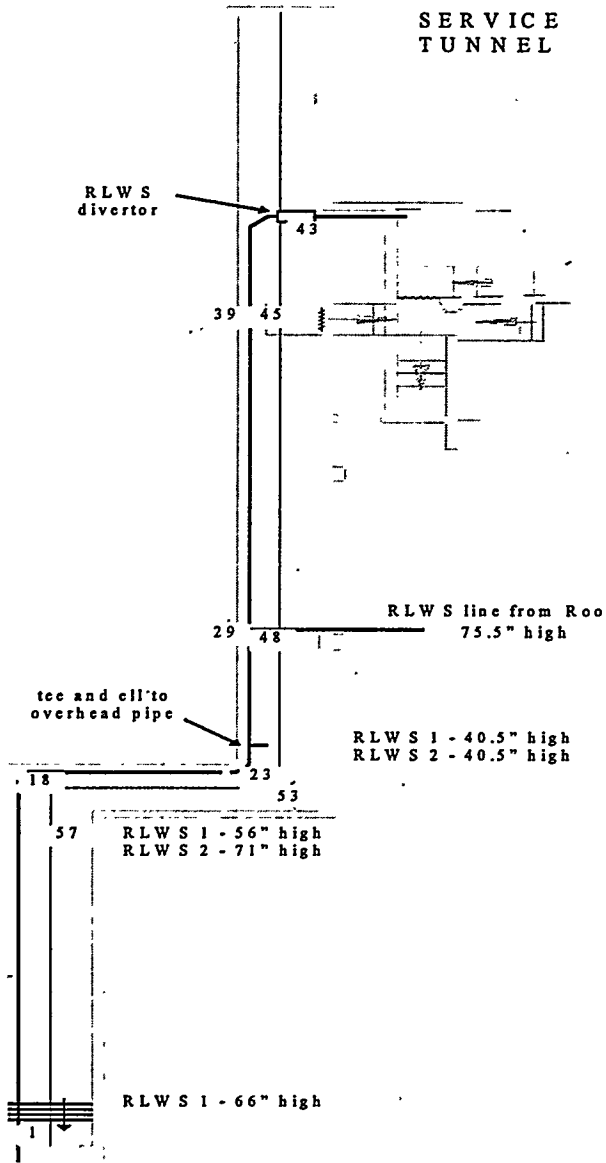


Measurement Locations

Locations	Bottom	Comments	Locations	Bottom	Comments
4A4_1	300 c/m	RLWS pipe at door to 4A	4A4_11	2500	RLWS line
4A4_2	500	air duct at 1' from door	4A4_12	2500	RLWS line near tee
4A4_3	500	RLWS & air duct	4A4_13	1800	RLWS line at stub
4A4_4	400	air duct	4A4_14	1800	RLWS line
4A4_5	300	RLWS line	4A4_15	200	decon line
4A4_6	200	RLWS line	4A4_16	800	plenum - upper half
4A4_7	400	RLWS line	4A4_17	400	plenum - angled
4A4_8	450	RLWS line	4A4_18	900	plenum - center of bottom
4A4_9	650	RLWS line	4A4_19	100	decon line
4A4_10	900	RLWS line near elbow	4A4_20	1800	duct on ceiling - center

Data Entered by: _____

Date: _____



Measurement Locations

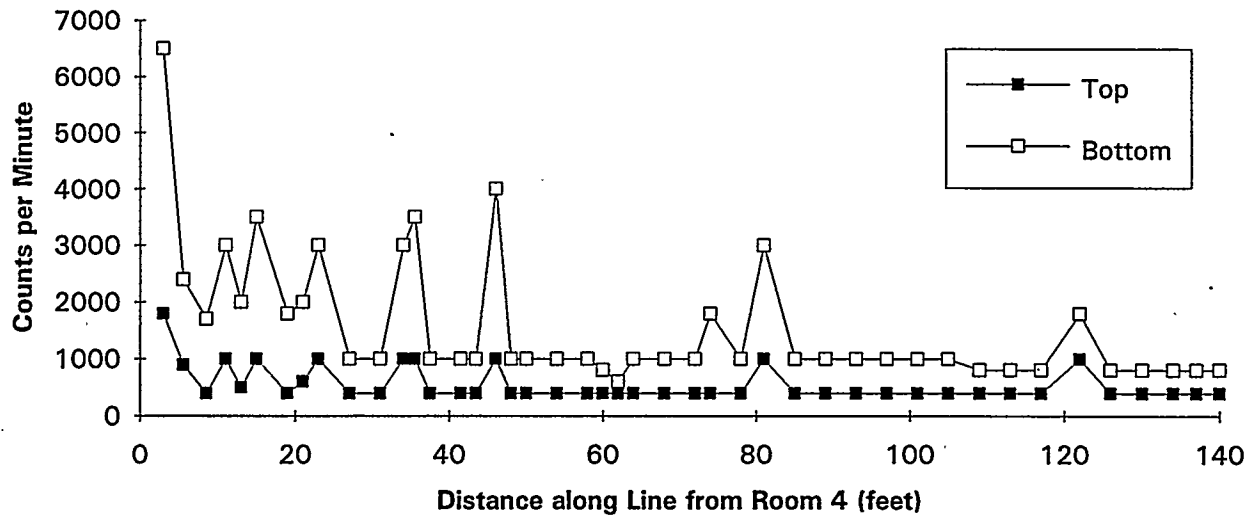
Locations	Top	Bottom	Comments
ST_1	1800 c/m	6500 c/m	RLWS line at 3' from wall to Room 4
ST_2	900	2400	+2.5' down pipe
ST_3	400	1700	+3'
ST_4	1000	3000	+2.5'
ST_5	500	2000	+2'
ST_6	1000	3500	+2'
ST_7	400	1800	+4'
ST_8	600	2000	+2'
ST_9	1000	3000	+2'
ST_10	400	1000	+4'
ST_11	400	1000	+4'
ST_12	1000	3000	+3' (weld)
ST_13	1000	3500	+1.5'
ST_14	400	1000	+2'

ST_15	400	1000	+4'
ST_16	400	1000	+2'
ST_17	1000	4000	+2.5'
ST_18	400	1000	+2' - bend in pipe
ST_19	400	1000	+2' from elbow
ST_20	400	1000	+4'
ST_21	400	1000	+4'
ST_22	400	800	+2'
ST_23	400	600	+2' - at elbow
ST_24	400	1000	+2'
ST_25	400	1000	+4'
ST_26	400	1000	+4'
ST_27	400	1800	+2' - at weld
ST_28	400	1000	+4'
ST_29	1000	3000	+3'
ST_30	400	1000	+4'
ST_31	400	1000	+4'
ST_32	400	1000	+4'
ST_33	400	1000	+4'
ST_34	400	1000	+4' - at weld
ST_35	400	1000	+4'
ST_36	400	800	+4'
ST_37	400	800	+4'
ST_38	400	800	+4'
ST_39	1000	1800	+5'
ST_40	400	800	+4'
ST_41	400	800	+4'
ST_42	400	800	+4' - at elbow
ST_43	400	800	+3'-3" at divertor box for RPS line
ST_44	400	800	+31" (or 70" to wall)
ST_45	200	200	+13' from tank pit divertor box
ST_46	200	200	+13' (in line with ST_36)
ST_47	200	200	+20'
ST_48	200	200	+9' (T and elbow at line to Room 18)
ST_49	200	400	+2' (in line with ST_28)
ST_50	20000	6000	+2' - hot spot
ST_51	10000	4000	+2' - hot spot
ST_52	300	200	+4'
ST_53	200	3500	+7' - elbow at corner
ST_54	100	300	+4' - pipe at bend
ST_55	375	500	+7'
ST_56	100	300	+4'
ST_57	200	400	+10'
ST_58	200	400	+20'
ST_59	200	300	+16' (10' to wall of Room 4)

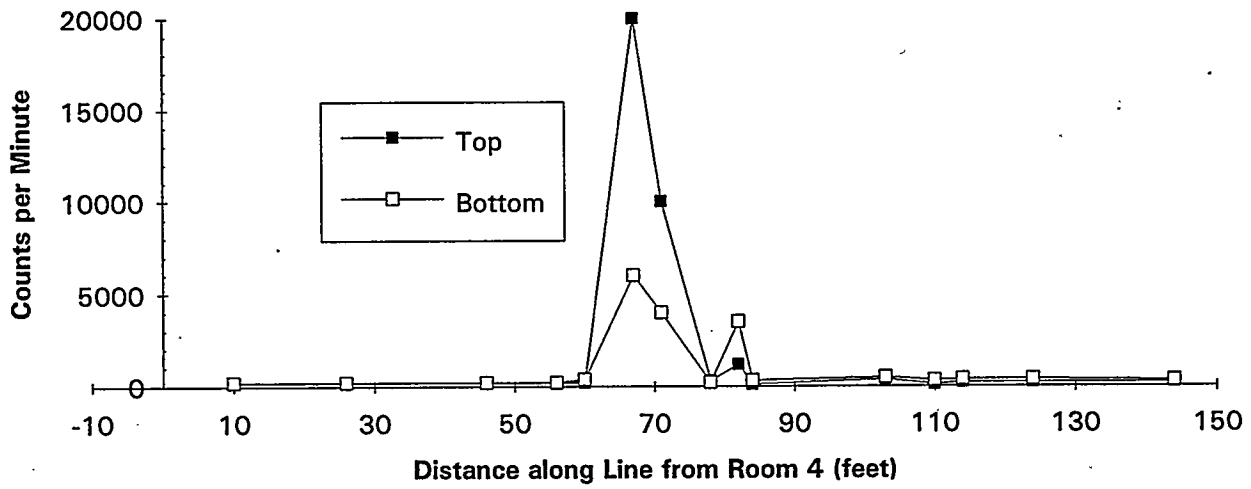
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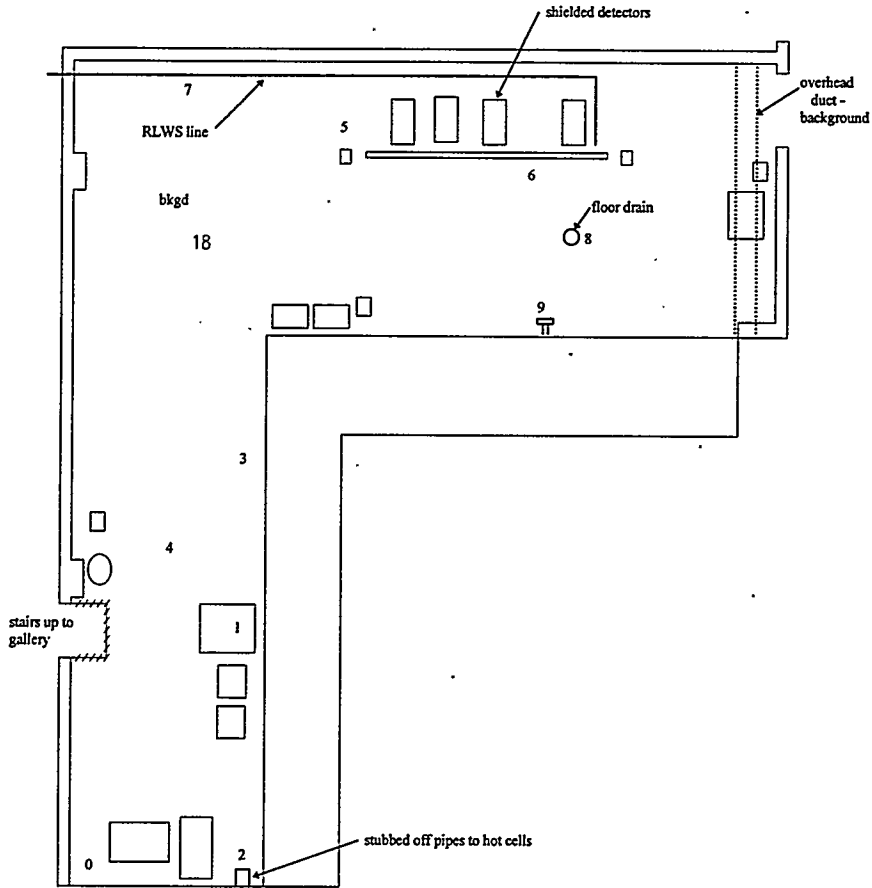
Date: _____

Radiological Survey of RLWS Line 1 in Service Tunnel



Radiological Survey of RLWS Line 2 in Service Tunnel



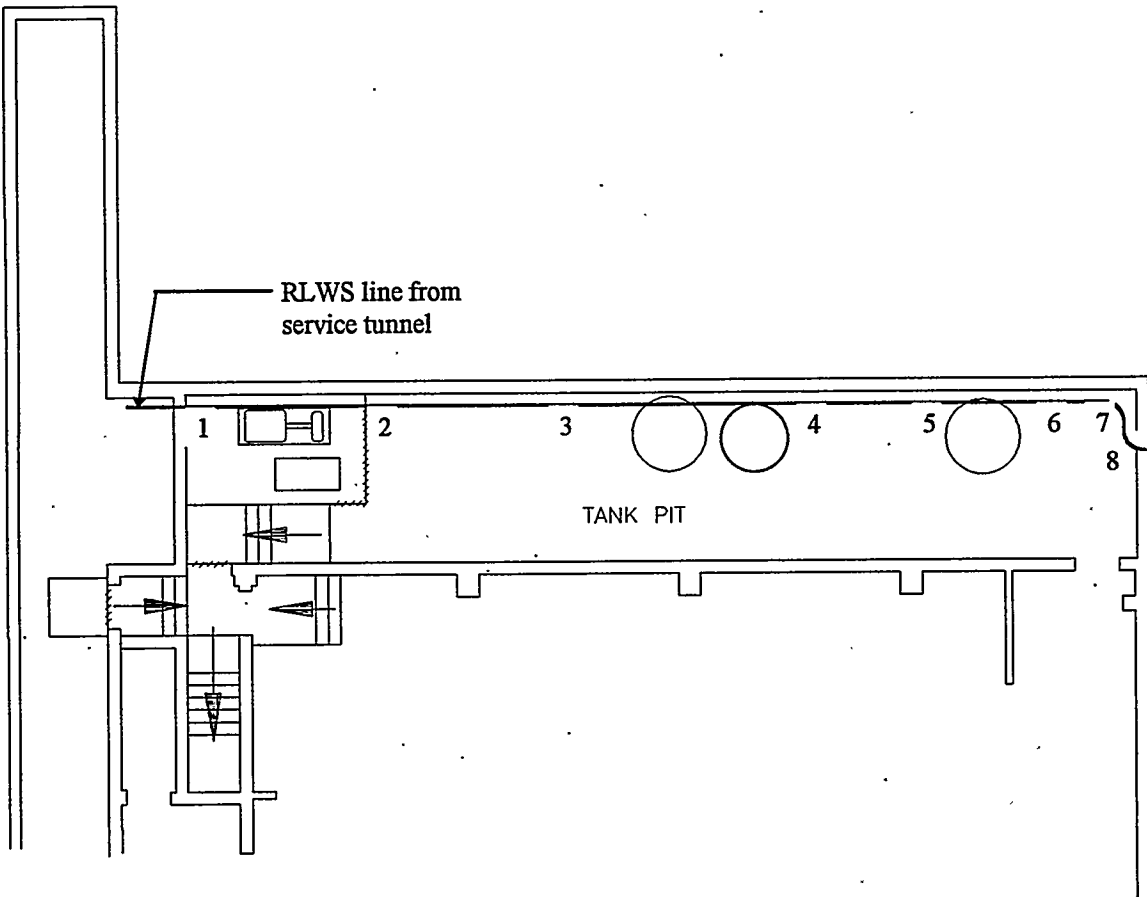


Measurement Locations

Locations	Top	Bottom	Comments
18_1	<u>700.mR/h</u>	<u>behind shield</u>	<u>Old furnace penetration - liquid leak</u>
18_2	<u>15 mR/h</u>	<u>(as posted)</u>	<u>5 capped pipes - penetrations into wall</u>
18_3	<u>140 mR/h</u>	<u>(as posted)</u>	<u>on wall at 11' to furnace penetration</u>
18_4	<u>800 c/m</u>	<u>background</u>	<u>floor drain</u>
18_5	<u>200 c/m</u>	<u>background</u>	<u>RLWS divertor</u>
18_6	<u>200 c/m</u>	<u>background</u>	<u>RLWS divertor</u>
18_7	<u>250 c/m</u>	<u>"hot spot"</u>	<u>RLWS line 5'8" from wall</u>
18_8	<u>1150 c/m</u>	<u>background</u>	<u>floor drain</u>
18_9	<u>2500 c/m</u>	<u>2500 c/m</u>	<u>crib waste penetration</u>

Data Entered by: _____

Date: _____

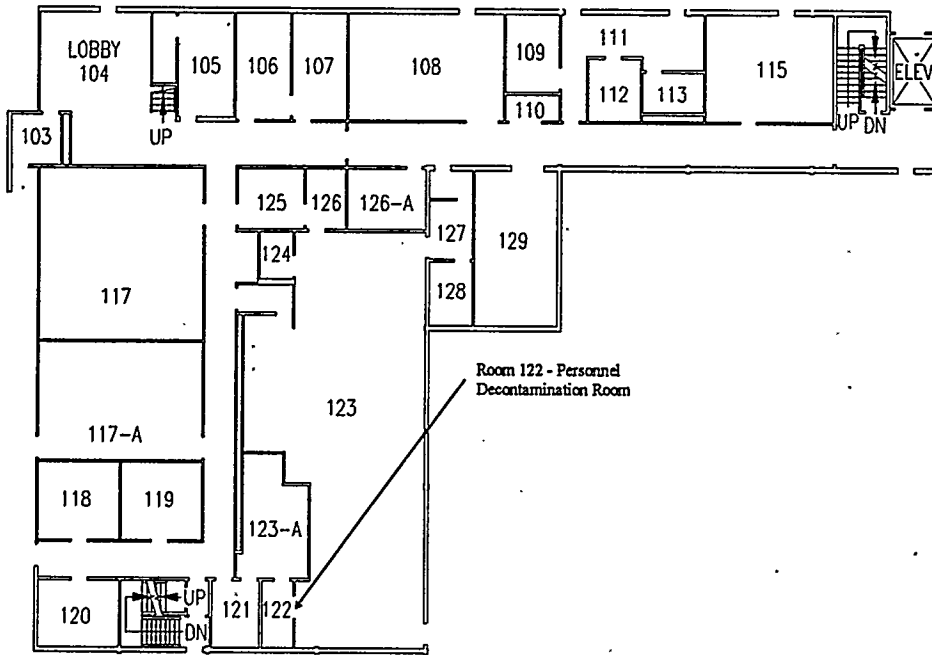


Measurement Locations

Locations	Top	Bottom	Comments
TP_1	<u>2000 c/m</u>	<u>2000 c/m</u>	<u>Stub 18" from wall</u>
TP_2	<u>400</u>	<u>400</u>	<u>12' from wall</u>
TP_3	<u>400</u>	<u>400</u>	<u>12' from TP_1</u>
TP_4	<u>400</u>	<u>400</u>	<u>16' from TP_2</u>
TP_5	<u>400</u>	<u>400</u>	<u>8' from TP_3</u>
TP_6	<u>400</u>	<u>400</u>	<u>12' from TP_4</u>
TP_7	<u>400</u>	<u>400</u>	<u>2' elbow</u>
TP_8	<u>800</u>	<u>800</u>	<u>6' from TP_6 at elbow - 2' to wall</u>

Data Entered by: _____

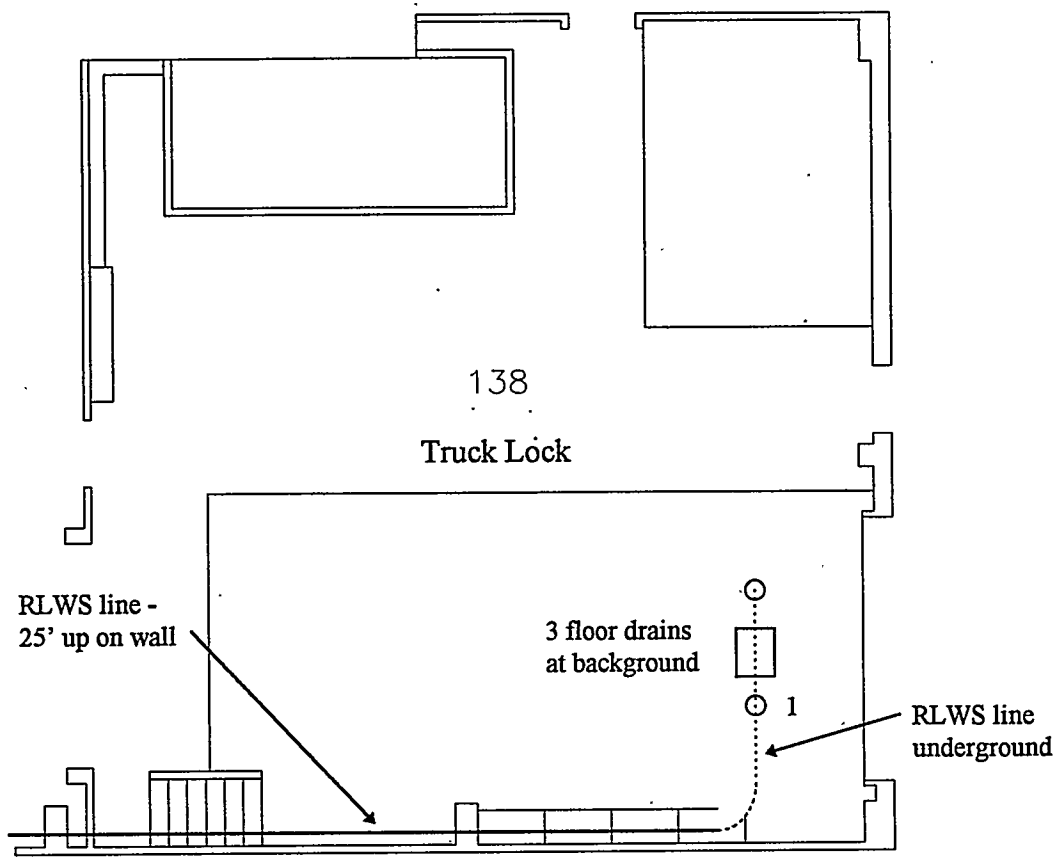
Date: _____



Measurement Locations

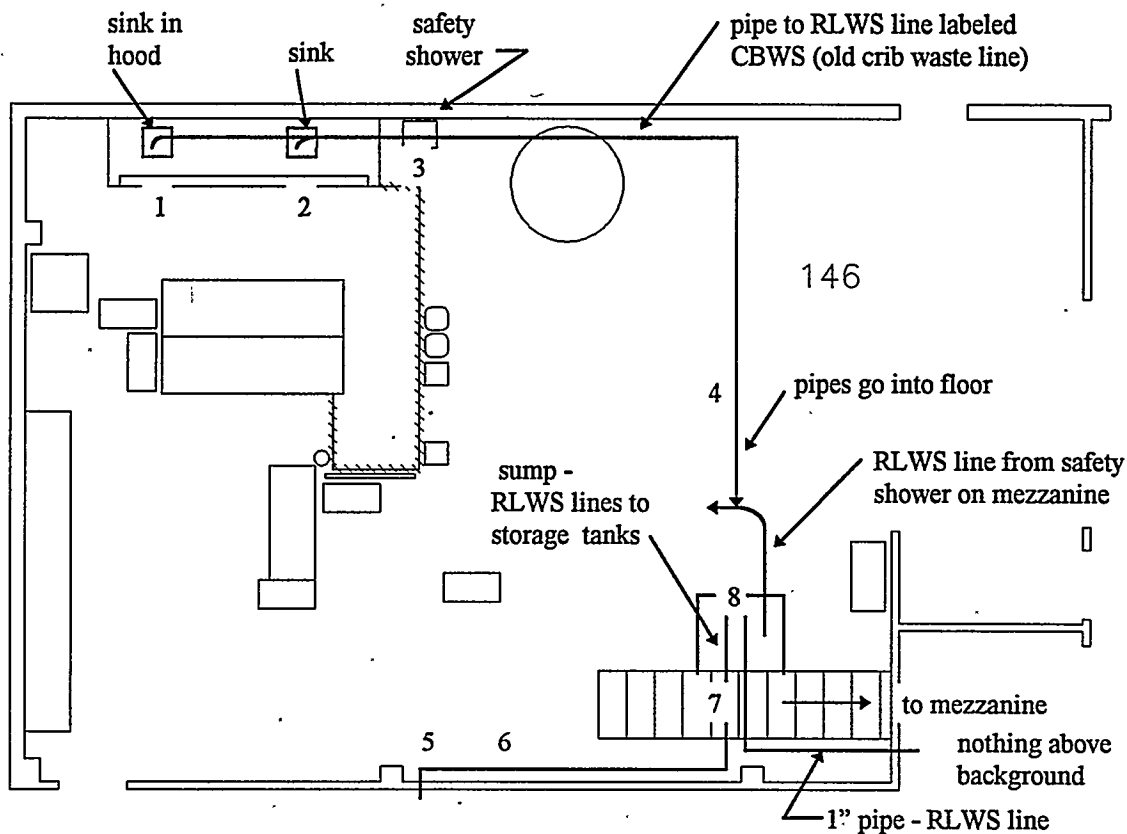
Locations	Top	Bottom	Comments
122_1	< 50 c/m	_____	<u>Shower drain</u>
122_2	<.50 c/m	_____	<u>Sink trap</u>
122_3	<50 c/m	_____	<u>RLWS line between sink and shower</u>

Data Entered by: _____ Date: _____



Measurement Locations

Locations	Top	Bottom
138_1	<u>background</u>	_____
138_2	_____	_____
138_3	_____	_____

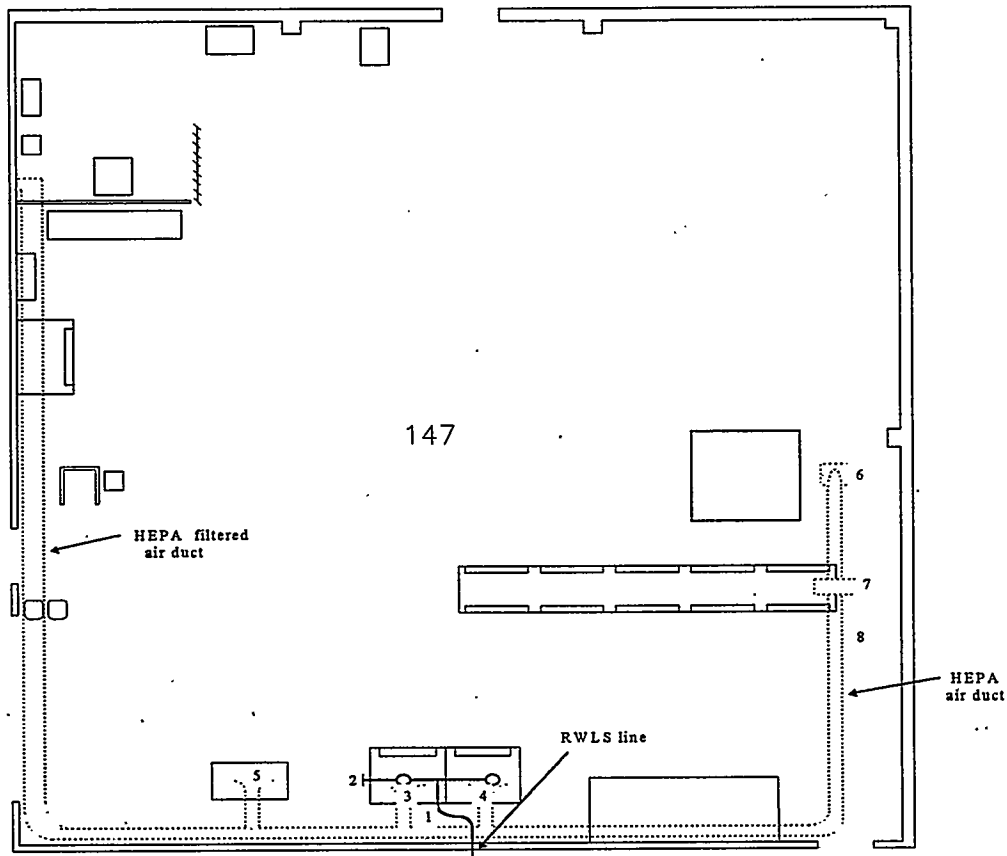


Measurement Locations

Locations	Top	Bottom	Comments
146_1	_____	<u>10,000 c/m</u>	<u>sink in hood (radium in trap) - RLWS</u>
146_2	_____	<u>background</u>	<u>sink in hood - pipe to RLWS line</u>
146_3	_____	<u>background</u>	<u>safety shower (mezzanine)</u>
146_4	_____	<u>background</u>	<u>trap into floor and sump - RLWS line</u>
146_5	<u>4000 c/m</u>	<u>4000 c/m</u>	<u>elbow of RLWS line from Room 147</u>
146_6	_____	<u>background</u>	<u>RLWS line from Room 147 - 2' from elbow</u>
146_7	_____	<u>background</u>	<u>RLWS line - middle of pipe under stairs</u>
146_8	_____	<u>high bkgd</u>	<u>maze of pipes in sump by stairs</u>

Data Entered by: _____

Date: _____



Measurement Locations

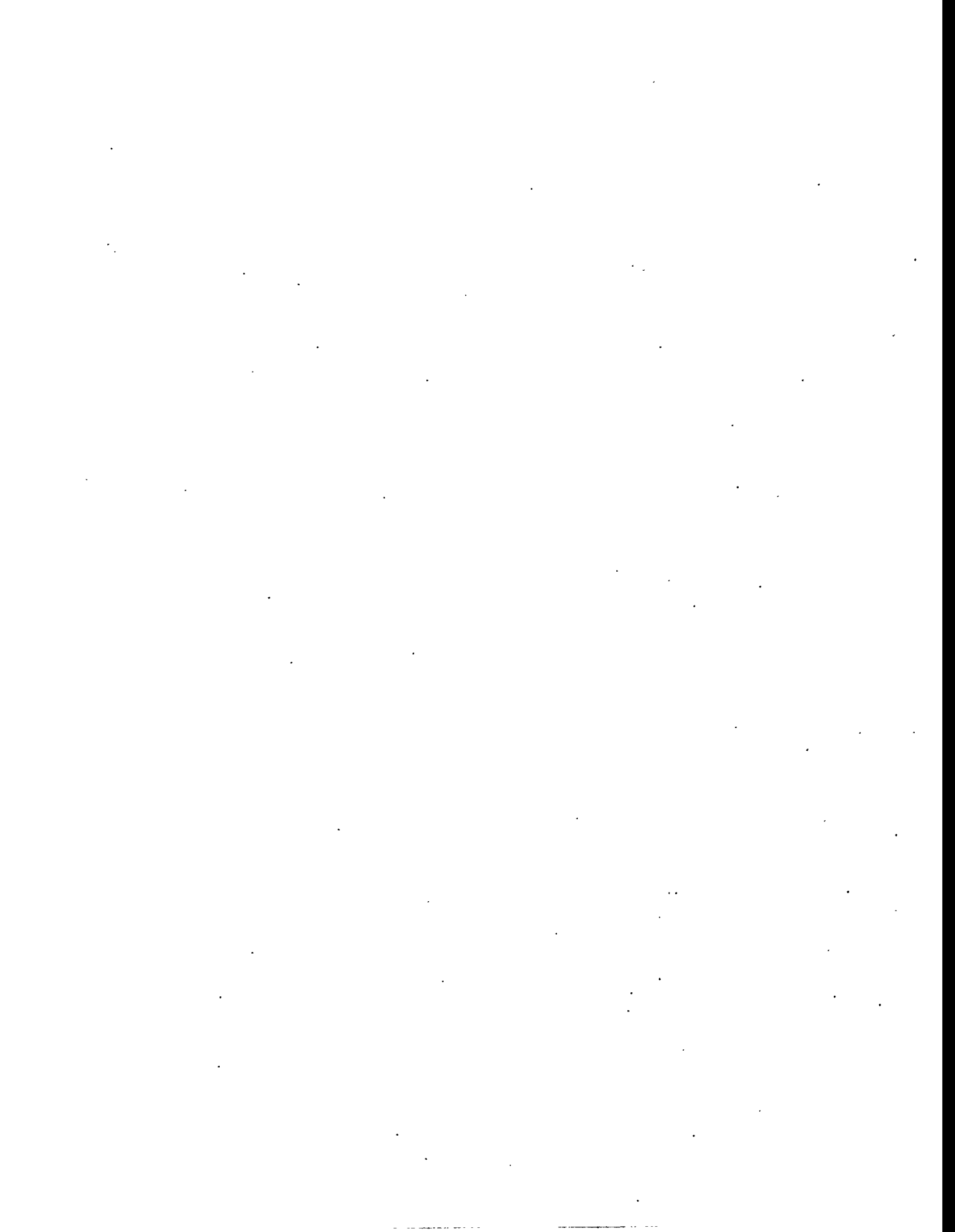
Locations	Top	Bottom	Comments
147_1	>10,000 c/m	_____	<u>liquid waste from decon hood</u>
147_2	high	_____	<u>decon hood RLWS pipe - cap on end</u>
147_3	high	_____	<u>duct to HEPA in decon hood</u>
147_4	>10,000 c/m	_____	<u>duct to HEPA in decon hood</u>
147_5	_____	_____	<u>duct to hood</u>
147_6	_____	_____	<u>elbow in duct from waste compactor</u>
147_7	_____	_____	<u>elbow in duct from manipulator box</u>
147_8	_____	_____	<u>elbow in duct past HEPA filter</u>

Data Entered by: _____

Date: _____

Appendix C

Neutron Measurement Results



Appendix C

Neutron Measurement Results

This appendix contains the spreadsheet analysis of the neutron counting data using the methods outlined in Section 4.2. Basically, the count rate is determined, then corrected for neutron background using the method explained in Section 4.2.2. Then, a correction is made to compensate for room scatter and distance from the source, using Equation 4.6. Finally, the background-corrected count rate is divided by the counts/second/gram of plutonium to derive the mass of plutonium.

Date	Location	Detector Orientation	Detector Ht.(in)	Det-Src Dist.(in)	ROI Start	ROI Stop	Integral Cts	Ct Time (sec)	Ct Rate (c/s)	Appropriate		Grams of Pu	Comments
										Bkgd	Bkgd		
3/28/95	3A-2	toward	18.5	24	93	2046	18659	300	62.20		21.42	3.86	RLWS pump
3/28/95	3A-2	away	18.5	24	93	2046	27678	300	92.26	81.55			
3/28/95	3A-3	toward	42.5	24	93	2046	19172	300	63.91		21.39	3.85	
3/28/95	3A-3	away	42.5	24	93	2046	28719	300	95.73	85.04			
3/28/95	3A-4	toward	10	12	93	2046	36010	300	120.03		53.87	3.01	
3/28/95	3A-4	away	10	12	93	2046	47778	300	159.26	132.32			
3/29/95	3A bkgd	Toward	18.5		93	2046	2391	100	23.91				vault door closed
3/29/95	3A bkgd	Toward	18.5		93	2046	2333	100	23.33				vault door closed
3/29/95	3A bkgd	Toward	18.5		93	2046	2366	100	23.66				vault door closed
3/29/95	3A bkgd	Away	18.5		93	2046	2255	100	22.55				vault door closed
3/29/95	Cf-1/324	Away	18.5	12	93	2046	16226	200	81.13				source check
3/29/95	Cf-1/324	Toward	18.5	12	93	2046	49564	200	247.82				source check
3/29/95	3A-1	Toward	63	36	93	2046	4178	300	13.93		-6.55		
3/29/95	3A-1	Away	63	36	93	2046	11305	300	37.68	40.96			pipe 82.5" above floor
3/29/95	3A-5	Toward	58.75	36	93	2046	13490	300	44.97				
3/29/95	3A-6	Toward	58.75	36	93	2046	10479	300	34.93				
3/29/95	3A-7	Toward	58.75	36	93	2046	7855	300	26.18				
3/29/95	3A-8	Toward	58.75	36	93	2046	6616	300	22.05				
3/29/95	3A-9	Toward	58.75	36	93	2046	5723	300	19.08				
3/29/95	3A-10	Toward	58.75	36	93	2046	4948	300	16.49				
3/29/95	3A-11	Toward	58.75	36	93	2046	4038	300	13.46			10.71	
3/29/95	3A Vault	toward	18.5	48	93	2046	70319	300	234.40				in front of vault door
3/29/95	3A Vault	toward	18.5	72	93	2046	38039	200	190.20				in front of vault door
3/29/95	3A Vault	toward	18.5	96	93	2046	45186	300	150.62				in front of vault door
3/29/95	3A Vault	toward	18.5	120	93	2046	35997	300	119.99				in front of vault door
3/29/95	3K-1	toward	65.5	36	93	2046	2079	300	6.93				Detector horizontal
3/29/95	3K-1	toward	77.5	24	93	2046	2265	300	7.55				Detector vertical
3/29/95	3K-1	toward	89.5	12	93	2046	2223	300	7.41		1.70	0.09	Detector vertical
3/29/95	3K-2	toward	89.5	12	93	2046	2137	300	7.12		1.41	0.08	
3/29/95	3K-3		93	24	93	2046	1916	300	6.39		0.68	0.12	
3/29/95	3K-bkgd		15.5	86	93	2046	1713	300	5.71				Under 3K-1
3/29/95	3K-4		56	24	93	2046	985	300	3.28		1.73	0.31	
3/29/95	3K-5	toward	56	24	93	2046	1151	300	3.84		2.47	0.44	
3/29/95	3K-5	away	56	24	93	2046	1191	300	3.97	2.74			Detector vertical
												1.05	

Date	Location	Detector Orientation	Detector Ht (in)	Det-Src Dist (in)	ROI Start	ROI Stop	ROI	Integral Cts	Ct Time (sec)	Ct Rate (c/s)	Appropriate Bkgd	Corr. for Bkgd	Grams of Pu	Comments
3/29/95	3J-21	toward	86	3.5	93	2046		1188	300	3.96		-0.51		Detector horizontal
3/29/95	3J-21bkgd		14	75	93	2046		1342	300	4.47				
3/30/95	Cf-1/324		12	12	93	2046		48419	200	242.10				source check
3/30/95	3Abkgd	toward	12					3376	100	33.76				
3/30/95	3Abkgd	toward	12					3245	100	32.45				
3/30/95	3Abkgd	away	12					2587	100	25.87				
3/30/95	3J-13	toward	77.5	24	93	2046		205	300	0.68	0.04	0.01		Detector flat
3/30/95	3J-13bkgd		15		93	2046		194	300	0.65				
3/30/95	3J-15	toward	60.5	24	93	2046		332	300	1.11	0.14	0.02		
3/30/95	3J-15bkgd		15		93	2046		291	300	0.97				
3/30/95	3J-11	toward	82	24	93	2046		153	300	0.51	0.06	0.01		
3/30/95	3J-11bkgd		15	24	93	2046		136	300	0.45				
3/30/95	3J-8	toward	81.5	24	93	2046		252	300	0.84	0.39	0.07		
3/30/95	3J-5	toward	75	30	93	2046		247	300	0.82	0.37	0.10		
3/30/95	3J-3	toward	82	24	93	2046		218	300	0.73	0.27	0.05		
3/30/95	3J-1	toward	81	24	93	2046		191	300	0.64	-0.01			background
3/30/95	3J-1bkgd		14	24	93	2046		193	300	0.64				
4/3/95	Cf-1/324		18.5	24	93	2046		30055	200	150.28		0.26		source check
4/3/95	3C-1	toward	18.5	36	93	2046		90	300	0.30	0.22	0.08		Distance to CL of hood
4/3/95	3C-2	toward	48	36	93	2046		123	300	0.41	-0.36	0.13		
4/3/95	3C-2	away	48	36	93	2046		83	300	0.28	0.10			
4/3/95	3B-1	toward	65	36	93	2046		252	300	0.84	0.68	0.24		CL of pipe 69"
4/3/95	3B-2	toward	65	36	93	2046		232	300	0.77	0.60	0.21		
4/3/95	3B-3	toward	65	36	93	2046		218	300	0.73	0.53	0.19		
4/3/95	3B-4	toward	65	36	93	2046		416	300	1.39	1.41	0.49		
4/3/95	3B-4	away	64	36	93	2046		196	300	0.65	-0.05			
4/3/95	3B-5	toward	43	36	93	2046		239	300	0.80	0.63	0.22		RLWS - jog in pipe
4/5/95	Cf-1/324		18.5	24	93	2046		31088	200	155.44		1.34		source check
4/5/95	Cf-1/324		18.5	24	93	2046		30625	200	153.13				source check
4/5/95	4A-21	toward	18.5	62	93	2046		164	300	0.55	-0.16			sump area
4/5/95	4A-19	toward	20	36	93	2046		173	300	0.58	-0.13			horizontal pipe
4/5/95	4A-18	toward	26	36	93	2046		217	300	0.72	0.49	0.17		bottom of duct

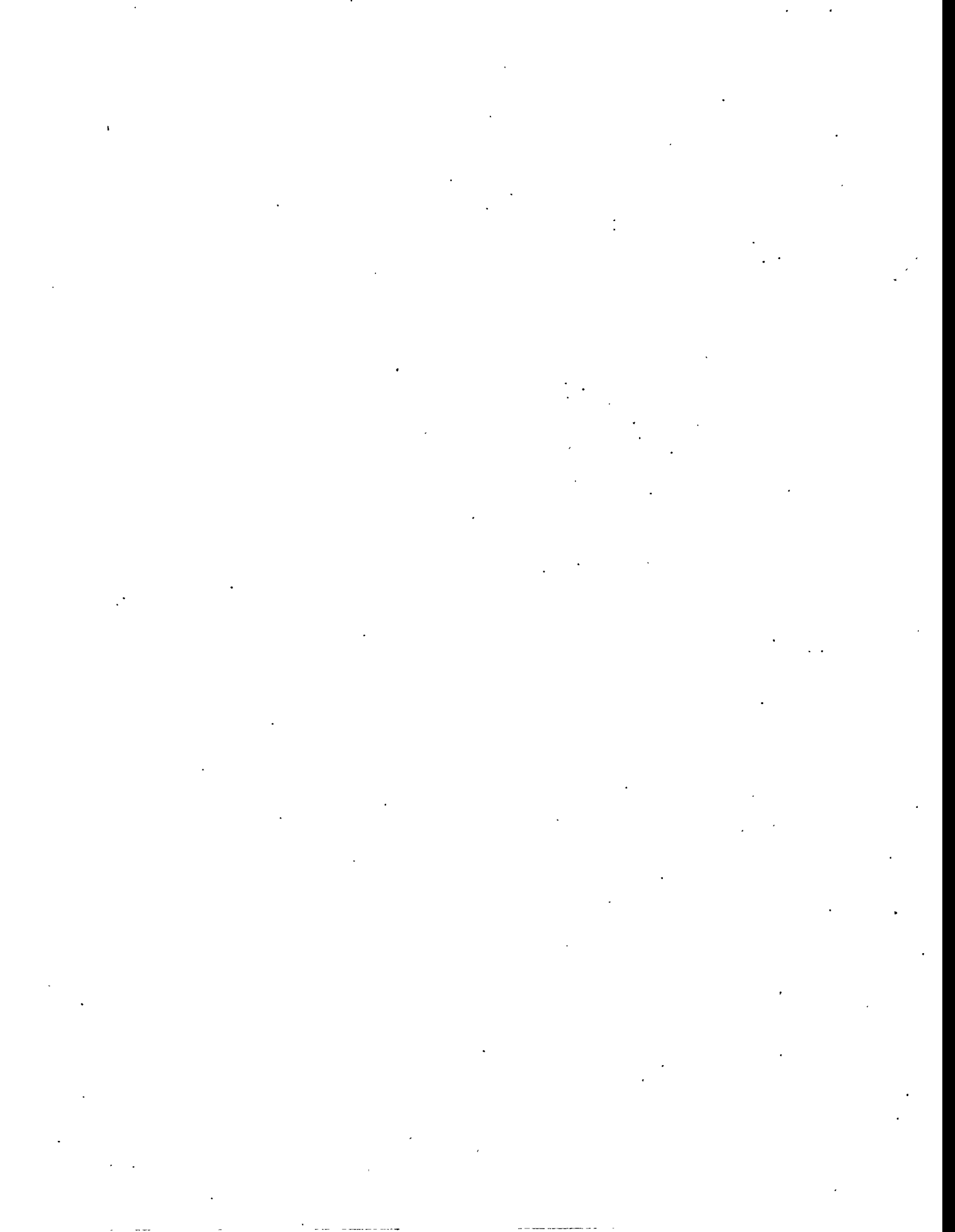
Date	Location	Detector Orientation	Detector Ht (in)	Det-Src Dist (in)	ROI Start	ROI Stop	Integral Cts	Ct Time (sec)	Ct Rate (c/s)	Appropriate Bkgd	Corr. for Bkgd	Grams of Pu	Comments
4/5/95	4A-17	toward	66	20	93	2046	242	300	0.81		0.60	0.08	to blue mark
4/5/95	4A-18	away	26	36	93	2046	212	300	0.71	0.46	-0.08		
4/5/95	4A-12	toward	52	36	93	2046	188	300	0.63		-0.08		closer
4/5/95	4A-12	toward	64	24	93	2046	187	300	0.62		-0.09		
4/5/95	4A-14	toward	64	24	93	2046	186	300	0.62		-0.08		
4/5/95	4A-8	toward	74	24	93	2046	237	300	0.79		0.08	0.02	
4/5/95	4A-3	toward	77	24	93	2046	448	300	1.49		-0.21		to blue on duct
4/5/95	4A-3bkgd	toward	15	87	93	2046	512	300	1.71				background
4/6/95	Cf-1/324		18.5	24	93	2046	30715	200	153.58			0.27	source check
4/6/95	ST-3	toward	66	12	93	2046	138	300	0.46		0.42	0.02	
4/6/95	ST-6	toward	64	12	93	2046	141	300	0.47		0.44	0.02	
4/6/95	ST-9	toward	64	12	93	2046	148	300	0.49		0.47	0.03	
4/6/95	ST-12	toward	63	12	93	2046	109	300	0.36		0.30	0.02	
4/6/95	ST-12	away	63	12	93	2046	85	300	0.28	0.14			
4/6/95	ST-13	toward	63	12	93	2046	127	300	0.42		0.38	0.02	
4/6/95	ST-17	toward	62.5	12	93	2046	97	300	0.32		0.24	0.01	
4/7/95	ST-20	toward	53	20	93	2046	113	300	0.38		0.27	0.04	
4/7/95	ST-53	toward	41	18	93	2046	105	300	0.35		0.24	0.03	center at *
4/7/95	ST-48	toward	20	12	93	2046	84	300	0.28		0.14	0.01	center at *
4/7/95	ST-48	away	20	12	93	2046	103	300	0.34	0.27			
4/7/95	ST-34	toward	30	36	93	2046	116	300	0.39		0.17	0.06	
4/7/95	ST-44	toward	18.5	12	93	2046	182	300	0.61		0.42	0.02	pipe CL 16" high
4/7/95	ST-44	away	18.5	12	93	2046	177	300	0.59	0.38			
4/7/95	ST-43	toward	22	12	93	2046	168	300	0.56		0.35	0.02	
4/7/95	ST-39	toward	19	30	93	2046	125	300	0.42		0.21	0.05	
4/7/95	ST-39	away	19	30	93	2046	156	300	0.52	0.42			
4/7/95	ST-50	toward	40	12	93	2046	96	300	0.32		0.20	0.01	
4/7/95	ST-51	toward	40	12	93	2046	96	300	0.32		0.20	0.01	
4/7/95	Cf-1/324	toward	18.5	24	93	2046	31425	200	157.13				source check
4/7/95	Cf-1/324	toward	18.5	24	93	2046	31751	200	158.76				source check
4/11/95	Cf-1/324		18.5	24	93	2046	30678	200	153.39			0.37	source check
4/11/95	3F-1	toward	54	12	93	2046	2008	300	6.69		7.52	0.42	detector facing up
4/11/95	3F-2	toward	66.5	12	93	2046	1391	300	4.64		2.53	0.14	detector facing up
4/11/95	3F-3	toward	18.5	24	93	2046	816	300	2.72		2.24	0.40	detector vertical
4/11/95	3F-3	away	18.5	24	93	2046	623	300	2.08	0.96			

Date	Location	Detector Orientation	Detector Ht (in)	Det-Src Dist (in)	ROI Start	ROI Stop	Integral Cts	Ct Time (ssec)	Ct Rate (c/s)	Appropriate Bkgd	Corr. for Bkgd	Grams of Pu	Comments
4/11/95	3F-3	side	18.5	24	93	2046	495	300	1.65				
4/11/95	3F-4	toward	18.5	36	93	2046	136	300	0.45		0.08	0.03	
4/11/95	3F-5	toward	18.5	36	93	2046	129	300	0.43		0.05	0.02	
4/11/95	3F-6	toward	18.5	38	93	2046	264	300	0.88		0.65	0.25	
4/11/95	3F-6	away	18.5	38	93	2046	236	300	0.79	0.46			
4/11/95	3F-1bkgd	away	16	64	93	2046	633	300	2.11			1.25	
4/11/95	3G-2	toward	16	24	93	2046	208	300	0.69		-0.09		detector horizontal
4/26/95	18-8	toward	14	12	93	2046	769	300	2.56		0.91	0.05	Floor Drain
4/26/95	18-4	toward	14	12	93	2046	1864	300	6.21		4.56	0.25	
4/26/95	18-2	toward	24	24	93	2046	63566	300	211.89				Stub centerline
4/26/95	18-2	toward	24	36	93	2046	52187	300	173.96				
4/26/95	18-1	toward	24	53	93	2046	4584	300	15.28		13.63	8.33	
4/26/95	18-3	toward	18.5	24	93	2046	901	300	3.00		1.35	0.24	
4/26/95	18bkgd	away	18.5		93	2046	496	300	1.65				Background
4/26/95	18-9	toward	18.5	12	93	2046	874	300	2.91		1.26	0.07	
4/26/95	18-5	toward	48	28	93	2046	572	300	1.91		1.44	0.33	
4/26/95	18-7	toward	49	24	93	2046	610	300	2.03		0.38	0.07	
4/28/95	18-0	toward	24	30	93	2046	2452	300	8.17		6.03	1.57	towards = East
4/28/95	18-0	north	24	30	93	2046	4630	300	15.43		17.01	17.17	Facing North
4/28/95	18-0	away	24	30	93	2046	2192	300	7.31	4.29			Away (west)
4/28/95	18-0	south	24	30	93	2046	1605	300	5.35	-3.16			Facing South
4/28/95	18-2	toward	24	48	93	2046	57228	300	190.76				4' to center of stubs
4/28/95	18-2	toward	24	36	93	2046	55106	300	183.69		227.37	79.21	3' to center of stubs
4/28/95	18-2	toward	24	24	93	2046	67832	300	226.11				2' to center of stubs
4/28/95	18-2	toward	24	72	93	2046	31318	300	104.39				6' to center of stubs
4/28/95	18-2	away	24	36	93	2046	7895	300	26.32	-87.37			away at 3' location
4/28/95	18-2	toward	48	36	93	2046	36023	300	120.08		142.56	45.40	2' above 5 stubs
												9.36	
5/23/95	147-5	Toward	108	34	93	2046	947	300	3.16		1.37	0.44	
5/23/95	147-5	Away	108	34	93	2046	1276	300	4.25	3.57			
5/23/95	147-1	Toward	19	51	93	2046	590	300	1.97		0.40	0.23	
5/23/95	147-1	Away	19	51	93	2046	998	300	3.33	3.12			
5/23/95	147-2	Toward	19	12	93	2046	613	300	2.04		0.09	0.00	
5/23/95	147-2	Away	19	12	93	2046	1186	300	3.95	3.91			
5/23/95	147-3	Toward	100.5	30	93	2046	769	300	2.56		1.33	0.35	

Date	Location	Detector Orientation	Detector Ht (in)	Detector Dist (in)	Det-Src Dist (in)	ROI Start	ROI Stop	ROI	Integral Cts	Ct Time (sec)	Ct Rate (c/s)	Appropriate Bkgd	Corr. for Bkgd	Grams of Pu	Comments
5/23/95	147-3	Away	100.5	30	93	2046			940	300	3.13	2.47			
5/23/95	147-4	Toward	104	34	93	2046			714	300	2.38		1.08	0.35	
5/23/95	147-6	Toward	60	36	93	2046			521	300	1.74		1.13	0.39	
5/23/95	147-6	Away	60	36	93	2046			535	300	1.78	1.22			
5/23/95	147-8	Toward	86	22	93	2046			578	300	1.93		1.51	0.23	Detector flat
5/23/95	147-7	Toward	87	19	93	2046			640	300	2.13		1.78	0.22	Detector flat
5/23/95	147-7&8	down	12	far	93	2046			478	300	1.59	0.70			Rm Bkgd - Det. flat/facing down
														2.21	
5/24/95	146-1	Toward	18.5	12	93	2046			494	300	1.65		1.11	0.06	distance to front of cabinet
5/24/95	146-1	Away	18.5	12	93	2046			489	300	1.63	1.08			"
5/24/95	146-2	Toward	18.5	12	93	2046			512	300	1.71		1.19	0.07	
5/24/95	146-3	Toward	8	18	93	2046			402	300	1.34		0.70	0.08	distance to drain
5/24/95	146-4	Toward	18.5	44	93	2046			785	300	2.62		2.02	0.95	
5/24/95	146-4	Away	18.5	44	93	2046			661	300	2.20	1.19			
5/24/95	146-5	Toward	12	2	93	2046			1049	300	3.50		2.92	0.01	
5/24/95	146-6	Toward	12	6	93	2046			1071	300	3.57		3.02	0.06	
5/24/95	146-6	Away	12	12	93	2046			785	300	2.62	1.11			
5/24/95	146-7	Toward	8	7	93	2046			801	300	2.67		1.39	0.03	
5/24/95	146-7	Away	8	7	93	2046			978	300	3.26	2.57			
5/24/95	146-8	Toward	2	20	93	2046			443	300	1.48		-0.06		det. face down on grating
5/24/95	146-9	Toward	2	20	93	2046			487	300	1.62		0.13	0.02	
5/24/95	146-8	Away	4		93	2046			915	300	3.05	3.08			
5/24/95	146 bkkgd	Toward	18.5		93	2046			1336	300	4.45				background at analyzer
5/24/95	146 bkkgd	Away			93	2046			706	300	2.35				(bottom of stairs)
														1.28	
6/26/95	TP-7	Toward	18.5	24	93	2046			156	300	0.52		0.26	0.05	
6/26/95	TP-7	Away	18.5	24	93	2046			196	300	0.65	0.52			
6/26/95	TP-6	Toward	18.5	41	93	2046			182	300	0.61		0.33	0.14	
6/26/95	TP-6	Away	18.5	41	93	2046			217	300	0.72	0.56			
6/26/95	TP-3	Toward	18.5	24	93	2046			225	300	0.75		0.50	0.09	
6/26/95	TP-3	Away	18.5	24	93	2046			227	300	0.76	0.51			
6/26/95	TP-2	Toward	18.5	24	93	2046			204	300	0.68		0.44	0.08	
6/26/95	TP-2	Away	18.5	24	93	2046			208	300	0.69	0.47			
6/26/95	TP-0	Toward	18.5	36	93	2046			182	300	0.61		0.34	0.12	
6/26/95	TP-0	Away	18.5	36	93	2046			209	300	0.70	0.52			
														0.47	

Appendix D

Gamma Measurement Results



Appendix D

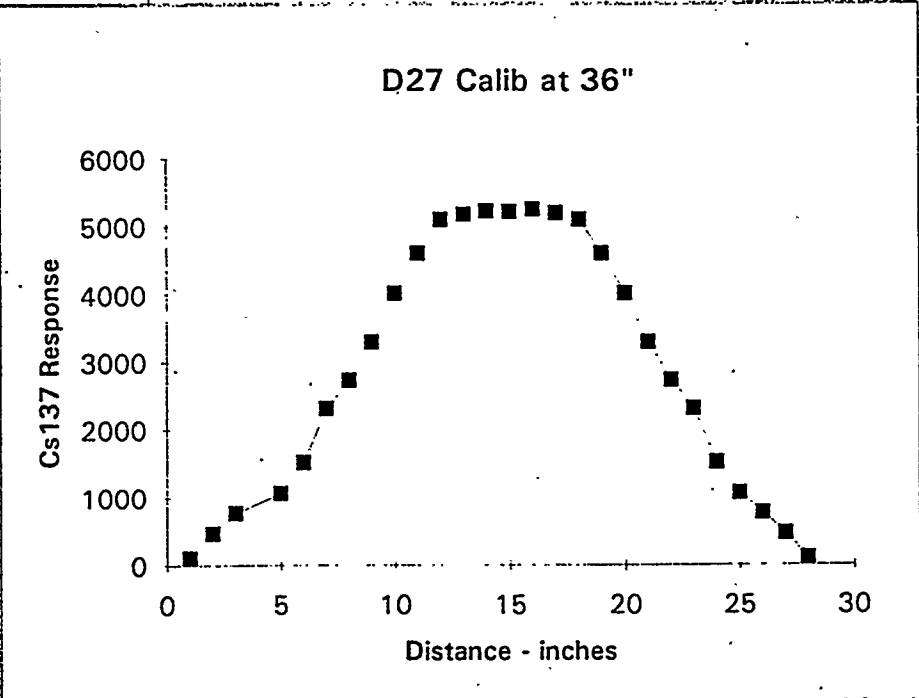
Gamma Measurement Results

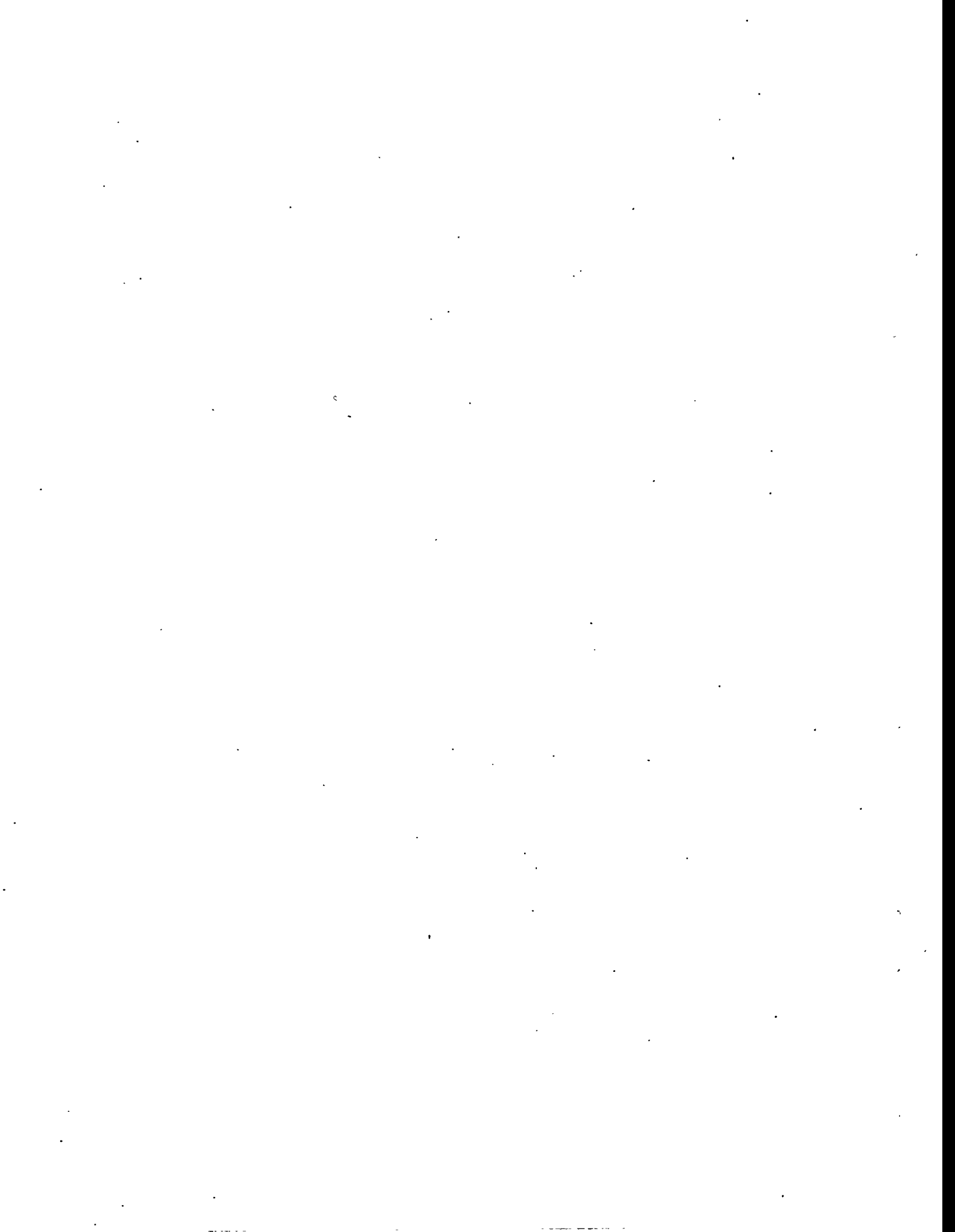
This appendix contains the results of gamma measurements along the radioactive liquid waste system (RLWS) pipeline that runs through the service tunnel in the basement, along the width of the 324 Building. The location numbers correspond to markings every 2 to 4 feet along the pipe where radiological survey measurements were performed. See page B.11 of Appendix B for the approximate location of the RLWS piping. Note that the zero position corresponds to the entrance to the service tunnel from room 4 in the southeast corner.

The figures show the curie content of the RLWS line given in Table 5.5. Almost all of the gamma activity is associated with ^{137}Cs , as shown in the first figure. The activity associated with other radionuclides is a factor of 100 times less, as shown in the second figure.

D27 FM Holdup Calibration at 36" 31-AUG-95								
				Ci	Ci ²	ai	cm ²	
Distance	662 i	i*s	cm	Net c rate	s ⁻²			
1	106	13	33.02	1.06	1.12	526.98	558.6	
2	461	12	30.48	4.61	21.25	486.44	2242.5	
3	774	11	27.94	7.74	59.91	445.90	3451.3	
5	1064	10	25.40	10.64	113.21	405.37	4313.1	
6	1519	9	22.86	15.19	230.74	364.83	5541.8	
7	2317	8	20.32	23.17	536.85	324.29	7513.9	
8	2736	7	17.78	27.36	748.57	283.76	7763.6	
9	3290	6	15.24	32.90	1082.41	243.22	8001.9	
10	4013	5	12.70	40.13	1610.42	202.68	8133.7	
11	4603	4	10.16	46.03	2118.76	162.15	7463.6	
12	5090	3	7.62	50.90	2590.81	121.61	6189.9	
13	5167	2	5.08	51.67	2669.79	81.07	4189.0	
14	5213	1	2.54	52.13	2717.54	40.54	2113.2	
15	5209	0	0.00	52.09	0.00	5.07	263.9	
16	5242	1	2.54	52.42	2747.86	81.07	4249.9	
17	5187	2	5.08	51.87	2690.50	121.61	6307.9	
18	5090	3	7.62	50.90	2590.81	162.15	8253.2	
19	4603	4	10.16	46.03	2118.76	202.68	9329.5	
20	4013	5	12.70	40.13	1610.42	243.22	9760.4	
21	3290	6	15.24	32.90	1082.41	283.76	9335.6	
22	2736	7	17.78	27.36	748.57	324.29	8872.6	
23	2317	8	20.32	23.17	536.85	364.83	8453.1	
24	1519	9	22.86	15.19	230.74	405.37	6157.5	
25	1064	10	25.40	10.64	113.21	445.90	4744.4	
26	774	11	27.94	7.74	59.91	486.44	3765.0	
27	461	12	30.48	4.61	21.25	526.98	2429.4	
28	106	13	33.02	1.06	1.12	567.51	601.6	
		14		779.64			149999.9	
L (cm) =	38.32564							
A(cm2) =	1479.247							
Kp =	0.000104	activity =	45.33966				Point source at 36"	
Kl	0.000249	activity =	108.1745				Line source at 36"	
Ka	0.000589	activity =	254.3574				Area source at 36"	
CF pipe =	1.26							

RLWS Line				uCi		
Location	Distance	Counts	TIME	Cs137		
22-44	36	11226	1000	29.32		
22-46	18	216	1000	0.14		
22-47	19	738	1000	0.54		
22-50	20	155564	1000	125.42		
22-51	20	89105	1000	71.84		
22-52	20	2170	1000	1.75		
22-53	24	17117	1000	19.87		
22-23	24	1192	1000	1.38		
22-20	24	721	300	2.79		
22-17	18	1098	300	2.39		
22-8	11	2559	300	2.08		
22-4	21.5	1886	300	5.86		
Room 3A						
Location	Distance	Counts	TIME	Cs137		
3A-1	33	120	1000	0.26		Duct
3A-2	36	360	300	3.13		RLWS
3A-3	36	142	300	1.24		RLWS
3A-TANK	36	149	300	1.30		
Room 3K						
Location	Distance	Counts	TIME	Cs137		
3K-1&2	36	3049	1000	7.96		duct
3K-4&5	36	858	1000	2.24		hood
Room 4						
Location	Distance	Counts	TIME	Cs137		
4-17	26	5446	1000	7.42		penion
4-18	36	19903	1000	51.99		penion
4-19	43	790	1000	2.94		RLW-
4-TANK	36	4826	1000	12.61		





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