2. SAMPLING

U.S. Department of Energy
201 Varick Street, 5th Floor
New York, NY 10014-4811

Page

2. SAMPLING

2.1 OVERVIEW

The purpose of environmental sampling and analysis is to obtain data that describe a particular site at a specific point in time from which an evaluation can be made as a basis for possible action. In this process, the collection of valid samples is the vital first step. Sampling should be done with the same care as the analysis, and both should be done with a rigor that is appropriate for the project at hand. In order for the data to be meaningful, sampling must be carried out with a clear purpose and with an understanding of the problem to be solved and the physical conditions that exist.

At EML, environmental sampling is carried out for purposes such as inventorying a pollutant at a specific point in time, calculating the pollutant transfer coefficients, and reconstructing deposition chronologies. Through these long-term studies, experience has been gained in sampling radioactive fallout, air particulates and gases, and total and rate of deposition. In this section, we describe the procedures developed by EML for environmental sampling. The corresponding analytical procedures are presented later in this Manual.

General guidance on collecting valid samples is given in Section 1.6.2. Unlike chemical or radiometric analyses, it is not possible to set down step-by-step procedures for sampling. For example, a variety of samples may be required for the purpose of establishing relationships between concentrations in different matrices to further the understanding of dynamic processes. Also, the concentration of a particular pollutant in an environmental matrix will change with time and location.

Usually, the crucial decisions in planning a sampling program are how many sites should be sampled and how often they should be sampled. These decisions can only be made based on a knowledge of the degree of variability due to these two factors (see Section 1.6.2). Most sampling programs require exploratory sampling so that the variability with time and location can be assessed in comparison with the required uncertainty. Experience has shown that statistical approaches based on these exploratory samples

usually lead to the taking of a smaller number of samples than would have otherwise been predicted. Another important consideration is that the number of samples must be consistent with the available analytical facilities.

Many times, the samples received in the laboratory may be representative of the particular conditions to be evaluated, but are not in the proper physical form for analysis. The samples may require reduction in size, drying or some form of homogenizing before subsamples can be taken for analysis. Some general considerations concerning sample preparation are discussed in Section 1.6.3 of this Manual.

The philosophy at EML is usually to collect a sufficient amount of sample so that there is not only enough to measure the constituent of interest, but also enough for reanalysis at a later time (see Section 1.6.3). Storage of samples for later analyses requires judgment in order to avoid loss of constituents to be measured or to avoid undesirable decomposition. EML maintains an extensive library of samples associated with its research programs, in some cases going back over 30 years (Klusek, 1989).

REFERENCE

Klusek, C. S.
"The EML Soil Inventory and Archive"
U.S. Department of Energy Report EML-519 (1989)

2.2 **AIR**

Contact Person(s) : Alfred Cavallo and Raymond J. Lagomarsino

2.2.1 **SCOPE**

Described in this section are the equipment and procedures used at EML in sampling for trace amounts of certain gases, liquids or solids dispersed in air. For gases, this involves either "whole air" samplers, or samplers which selectively adsorb the gas of interest. For the liquids and solids (aerosols), the techniques involve separating the particles from air by means of filtration or impaction.

In situ methods in which the sampling and analysis are one combined operation are not described as they are not commonly used at EML. Further information can be found in Air Sampling Instruments for Evaluation of Atmospheric Contaminants, a handbook published by the American Conference of Government Industrial Hygienists (ACGIH, 1989). This handbook, which is updated every 5 to 6 years, includes descriptions of practically every commercially available instrument for sampling trace gases and aerosols. It also has concise theory sections covering basic physics and experimental design pertinent to air sampling.

The locales for sampling in EML programs range from indoor (residential or occupational) to outdoor (surface to upper troposphere).

2.2.2 **AEROSOLS**

2.2.2.1
INTRODUCTION

Many of EML's research programs require that samples be taken of aerosols, defined as "a system of colloidal particles dispersed in a gas". The suspending gas is normally indoor or outdoor air. Smoke and mist are common examples of aerosols, but frequently

the particles are so small that the aerosol cannot be detected by eye. Typically, environmental aerosols contain a broad mixture of chemical species, both liquid and solid, including some radioactive materials.

The sampling of aerosols involves a number of complications that are not present when sampling for environmental gases. Therefore, specialized training or experience is highly desirable for personnel responsible for aerosol sampling. Some universities — such as New York University (Institute of Environmental Medicine) — conduct annual or semiannual one-semester courses in aerosol science; inquiries can be made about auditing such courses. Also, the University of Minnesota offers a 4-day short course each summer.

Excellent books are now available which consolidate the advances in aerosol science over the past 25 years. Two of the best are the textbook by Hinds (1982) and the handbook by the ACGIH (1989), already mentioned.

There are two broad categories of aerosol sampling methods: integral and size-selective. In the former, the goal is to collect a single sample in which the sizes and types of collected particles accurately represent those in the air. The integral samplers commonly used at EML are described in Sections 2.2.2.2 and 2.2.4. Size-selective samplers are designed to provide information on particle size as well as particle amount. Sections 2.2.2.3 through 2.2.2.8 describe the size-selective samplers commonly used at EML.

Among the size-selective samplers, the high volume cascade impactor and the diffusion battery (when supplemented with the appropriate chemical analysis) produce sufficient data to generate complete particle size spectra for the chemical species of interest. Although shortcuts are possible, the proper way to generate these spectra is to apply a suitable mathematical technique to "unfold" or "deconvolute" the data (see Section 2.2.2.8).

The modern method of presenting aerosol size spectrum data is the "generalized histogram" described in Chapter E of ACGIH (1989).

REFERENCES

Hinds, W. C.

"Aerosol Technology: Properties, Behavior and Measurement of Air borne
  Particles"

Wiley-Interscience, New York (1982)

Air Sampling Instruments for Evaluation of Atmospheric Contaminants

Hering, S. (Editor)

American Conference of Governmental Industrial Hygienists (ACGIH), Cincinnati, OH,
Seventh Edition (1989)

2.2.2.2
TOTAL PARTICLE COLLECTIONS

A. Introduction.

The procedures discussed in this section are limited to the specific requirements of programs currently conducted at EML and may not be applicable to other studies that require the total collection of air particles.

Since January 1963, EML has conducted the Surface Air Sampling Program (SASP). In 1987, the Remote Atmospheric Measurements Program (RAMP) was initiated as an extension and modification of SASP. The primary objectives of SASP/ RAMP are to identify and study the temporal and spatial distribution of anthropogenic and natural radionuclides in the lower troposphere. These objectives are achieved by filtration of large volumes of air to concentrate the radionuclides in the aerosol prior to sample analyses. The sampling procedures used in SASP/RAMP would not be appropriate if one requires particle size analyses or particle specific activity distributions. The filters used in these programs also will not collect uncharged, unattached radioactive gas molecules.

To sample large volumes of air and obtain total particle collection, it is necessary to use an appropriate filter material and an air mover. In SASP/RAMP, the air mover must be capable of continual operation at high flow rates under harsh environmental conditions. To calculate air concentrations of radionuclides, it is necessary to accurately determine the total volume of the air which has been sampled.

B. Filter material for total particle collections.

The criteria for filter selection are good collection efficiency for submicron particles at the range of face velocities used, high particle and mass loading capacity, low-flow resistance, low cost, high mechanical strength, low-background activity, compressibility, low-ash content, solubility in organic solvents, non-hygroscopicity, temperature stability, and availability in a variety of sizes and in large quantities. In the selection of a filter material, a compromise must be made among the above criteria that best satisfies the sampling requirements. An excellent review of air filter material used to monitor radioactivity was published by Lockhart et al. (1964). Lippmann (1989a) also provides information on the selection of filter materials for sampling aerosols by filtration.

Two filter sizes are presently used in SASP/RAMP, a 20.32 cm circle and a 20.32 cm × 25.40 cm rectangle. The polypropylene fiber filter, Dynaweb Grade DW7301L (see Specification 7.17) is used at all SASP and RAMP sites. The filter is composed of a 100% polypropylene web that is 100% binderless. Three layers of this web are collated and sandwiched between two sheets of a protective DuPont Reeme (100% polyester) scrim. The top scrim is removed prior to sampling at RAMP sites because after their return to EML for analysis these samples are compressed into pellets and the scrim hinders compression. At all other sites, the filter can be used during sampling with both top and bottom scrim in place.

C. Air movers.

A large variety of air movers are commercially available and have been reviewed by Rubow and Furtado (1989).

Many factors must be considered when selecting an air mover. Such factors as portability, power requirements, maximum operational flow rate/temperature/pressure, cost, durability, and maintenance must be considered in the selection of an air mover. It is also important that the air mover itself is not a source of contamination in any study.

The major factors that were considered in the selection of an air mover for the SASP sites were durability, low maintenance, and a flow rate of $\sim 1 \text{ m}^3 \text{ min}^{-1}$ [288°K, 101.3 kPa

(760 mm Hg)] at pressure drops across the filter ranging from ~ 5 kPa to ~ 20 kPa (20-80 in water).

Two air mover systems are currently used in SASP/RAMP. The SASP sites are equipped with a Roots Blower (see Specification 7.18) connected to a 1 HP electric motor (see Specifications 7.18) by a fan belt. The RAMP sites are equipped with a Fuji ring compressor (see Specification 7.18) in which the air mover is directly connected to a 0.5 HP electric motor. The Roots system must be enclosed in a louvered shelter to protect the pump from direct exposure to precipitation. The Roots system is frequently mounted on a 1-m high metal stand. The Fuji system is enclosed in a custom manufactured aluminum container. The Roots system is heavier than the Fuji system and may be more durable for continual operation in harsh environments. The Roots system exhausts some oil vapors which may be a contaminant in certain studies. The Roots system is designed for 20.32 cm round filters, while the Fuji system is designed for 20.32 cm $\times$ 25.40 cm rectangular filters.

D. Flow calibration and sample volume determination.

To calculate the concentrations of radionuclides in the lower troposphere or concentrations of any air pollutant collected and concentrated by filtration, it is necessary to accurately determine the total volume of air sampled.

Generally, a parameter of the air mover can be related to flow. If the mean flow during a collection period can be determined, the total volume of air sampled can be calculated. Accurate flow measurements and the total integrated sample volume of air can be obtained using a mass flow meter and a totalizer. This direct technique of air flow measurement becomes impractical at remote field locations due to cost and exposure of the flow meter to harsh environments. Other procedures for the measurement of air flow in sampling systems are reviewed by Lippmann (1989b).

We have determined that the best technique to measure flow, at sites equipped with Roots systems, is to determine an empirical relationship between the Roots blower inlet pressure and the flow through the upstream filter. An orifice meter was manufactured at EML to derive this relationship. The orifice meter has been calibrated for flow using secondary flow measurement devices that are traceable to a primary standard volume

meter. All Roots systems are calibrated using this orifice meter prior to field installation. Periodic calibrations are conducted at the field sites.

During sampling, the pressure at the Roots blower inlet is measured using a Magne-helic gauge. The initial and final pressure values are averaged. The average inlet pressure is then used in the previously described empirical relationship to calculate the average flow. This average flow is multiplied by the collection interval to obtain the total volume sampled. The total volume sampled is adjusted to a pressure of 101.3 kPa (760 mm Hg) and a temperature of $288^{\circ}K$.

At sites equipped with a Fuji system, the flow is determined using an empirical relationship between the pressure differential across a fixed orifice located in the blower exhaust. The initial and final pressure values are used to calculate a mean flow and a total sample volume as previously described. The relationship between flow and the pressure differential across the fixed orifice plate and in the Fuji flow system was determined using a Hastings Laminar flow element traceable to a primary standard volume meter.

REFERENCES

Lippmann, M.
"Sampling Aerosols by Filtration"
Hering, S. (Technical Editor)
in: Air Sampling Instruments, 7th Edition, American Conference of Governmental
  Industrial Hygienists, Cincinnati, Ohio, pp. 305-336 (1989a)

Lippmann, M.
"Calibration of Air Sampling Instruments"
Hering, S. (Technical Editor)
in: *Air Sampling Instruments,* 7th Edition, American Conference of Governmental
  Industrial Hygienists, Cincinnati, Ohio, pp. 73-100 (1989b)

Lockhart, L., R. Patterson and W. Anderson

"Characteristics of Air Filter Media Used for Monitoring Airborne Radioactivity"

Naval Research Laboratory Report NRL-6054, June (1964)

Rubow, K. and V. Furtado

"Air Moves and Samplers"

Hering, S. (Editor)

in: *Air Sampling Instruments,* 7th Edition, American Conference of Governmental
  Industrial Hygienists, Cincinnati, Ohio, pp. 241-274 (1989)

2.2.2.3

DICHOTOMOUS SAMPLER

A Sierra Model 245 automatic dichotomous sampler is used to study the "inhalable" (<10 µm) particles in ambient air. The original commercial design was developed and sold by Sierra Instruments Inc., but is now sold by Andersen Instruments, Inc., Sierra Anderson Division, 4801 Fulton Industrial Blvd., Atlanta, GA 30336.

The dichotomous sampler is capable of separating the particles <10 µm (determined by the aerosol 10 µm inlet cut-off size) into two fractions. By means of virtual impaction, the sampled particles are separated into two fractions, the fine fraction, <2.5 µm, and the coarse fraction between 2.5 and 10 µm. This technique has the added advantage of eliminating problems associated with particle bounce and re-entrainment that are sometimes experienced in cascade impactor sampling.

The particles are collected on Teflon membrane filters which are ideal for gravimetric analyses of the fine, coarse, and inhalable (sum of fine and coarse) fractions, and for chemical analyses by X-ray fluorescence or other high-resolution chemical techniques.

The experimental procedures used at EML are defined in the instrument manual along with literature references on the development and application of the sampler for studies of PM-10 compliance monitoring, source discrimination, fine-particle monitoring, and visibility monitoring.

2.2.2.4

HIGH VOLUME CASCADE IMPACTORS

A. Introduction.

A description of the Sierra Model 235 multistage cascade impactor, which is used to measure the particle size distribution of both indoor and outdoor aerosols, is presented here. The original design was developed and sold by Sierra Instruments, Inc., but now is sold by Andersen Instruments, Inc., Sierra Anderson Division, 4801 Fulton Industrial Blvd., Atlanta, GA 30336. A number of articles are available in the literature that deal with both the theoretical and experimental development of inertial impactors (Marple et al., 1973, 1974; Marple and Willeke, 1979).

At EML, this impactor is used for research studies and is not commonly used for long-term sampling projects. The methods described herein are used for 1-5 day sampling periods, and the units are checked on a daily basis.

B. Operating conditions.

The sampler is capable of sampling at flow rates ranging from 0.56 m^3 min^{-1} to 1.68 m^3 min^{-1}. The flow is regulated by a constant flow controller that automatically adjusts to any flow rate change caused by loading conditions or motor variances. In the majority of our outdoor sampling efforts, the unit is operated at a flow rate of 0.85 m^3 min^{-1} (30 cfm) to prevent overloading and to assure that the flow rate remains constant during the sampling interval.

Configuration. The unit is housed in a standard high-volume sampling shelter without a size selective inlet. Under some conditions a 10 µm or 15 µm size-selective inlet is used. The impaction substrate is slotted Whatman No. 41 filter paper treated with light mineral oil (Fisher Scientific, 50 Faden Road, Springfield, NJ 07081). The oiled filter minimizes particle rebound and reentrainment, which has been found during previous impactor evaluations to bias impactor size distribution measurements toward smaller sizes (Knuth, 1979a). This method of oil treating the filter is not compatible with mass determination by weighing methods. All of our analyses are done by chemical or radiochemical procedures.

General maintenance. The impactor plates are washed with alcohol and the slots are cleaned using a cotton swab or soft cloth. Care must be taken so that the integrity of the slot is not altered by using any cleaning material that may damage the slot spacing or edges in any manner. The plates are dried with compressed air and the slots are visibly examined to assure that no foreign material remains in the slot. The impactor plates should be numbered 1-5 in a corresponding corner of the plates, starting with the largest slotted plate numbered 1 to the smallest plate numbered 5.

Loading procedures. The loading and unloading of the impactor assembly should be done in a clean environment (clean room or clean bench). The loaded unit can then be transported to the sampling area and installed on site. In handling the filters, reasonable care should be taken to minimize contamination of the sample. Filters should be handled at the extreme corners, and latex gloves or forceps should be used during the following loading procedures.

1. Soak the total number of filter impaction substrates needed for the number of impactors being loaded in mineral oil using a Pyrex or comparable dish. Note: A few extra oiled filters may be required for blank analysis values.

2. After the filters are completely wetted, they are removed from the oil and allowed to drip until all excess oil has run off.

3. Starting with the slotted base plate, place a mineral oil wetted filter on the plate assuring that the paper is centered and all slots are open. Using a glass rod, assure that the paper is in contact with the plate and no air bubbles are present.

4. Carefully place the slotted plate, #5, on top of the filter. Assure that it is flat, and that it secures the impaction filter #5 beneath it to the base plate. At this time, the filter paper should be visible through the slots. If not, remove plate #5 and reposition it 180 degrees. Check again.

5. Continue placing filters and plates #4 to #1 as described in Step 4 above.

6. After positioning slotted plate #1, tighten the two screws that secure the five plates sandwiched to the base plate. At this time, wipe any excess oil from the sides of the

plate assembly. Note: If there is an extreme amount of oil seeping through the edges of the plates, start over at Step 1 allowing a longer time for the filters to drip dry.

7. The extra wetted filter is carefully folded over on itself once or twice and placed in a polyethylene bag. Attach an appropriate identification label to the bag.

8. The loaded impactor assembly is taken to the sampling site and placed in position over an appropriate back-up filter. For our work, a suitable back-up filter is determined dependent on the analytical procedure to be used. Back-up filters which have been used for different applications are glass fiber, Microsorban, Microdon, and Whatman No. 41.

9. Tighten wing nut assemblies on four corners of the base plate to secure impactor assembly to back-up filter holder.

Sampling procedure.

1. Start unit and adjust the in-line orifice reading to the desired setting. This setting corresponds to the flow rate determined from a calibration curve of the orifice.
   Note: A top loading calibrated orifice can be used to verify flow rate in the field.

2. Record all pertinent flow and time information required. Note: If a number of runs will be required for the experiment, a data sheet form should be developed and used for recording data.

3. Whenever possible, the unit should be visibly checked during the sampling period. If an in-line orifice is being used, the manometer reading can be read and recorded at any time during the exposure period without interrupting the sampling procedure.

4. At the end of the sampling period, record all data and if possible recheck the flow rate with the top loading orifice.

Sample recovery.

1. Carefully remove the impactor assembly from above the back-up filter. Immediately fold the back-up filter over on itself, soiled side in, twice if required, and place in prelabeled polyethylene bag.

2. Take the impactor assembly to a clean environment and begin the disassembly procedure by carefully removing the top plate, #1, folding the soiled filter over on itself, soiled side in, and placing it in a prelabeled envelope or polyethylene bag. Note: We have found that polyethylene bags are better than glassine envelopes for storing the oiled filters prior to analysis. A glass rod can be used for creasing the oiled filter when folding.

3. Place the five oiled impaction filter polyethylene bags into a larger polyethylene bag, appropriately labelled, along with the back-up filter and the appropriate blank oiled filter. This is the extra-oiled impaction filter prepared prior to loading the impactor. Note: Although you can usually determine the stage the oiled filter came from by visual determination of the width of the deposit on the filter, care should be taken when removing the filters from the impactor to assure that the filter is correctly identified when placed in the polyethylene bag.

4. The impactor plates can be placed in an alcohol bath to remove any excess oil from the plates prior to cleaning for assembly. Compressed air can be used to clean slots if necessary.

Data analysis. Various methods of describing the particle size distribution of sampled aerosol have been used through the years for impactor data. In order to use any one or more of these methods it is necessary to determine the amount of material collected on each of the impactor stages and back-up filter. At EML, this is usually done by measuring the radioactivity or by chemical analyses.

Two of the most used methods of describing the distribution derived from impactor data are histograms and cumulative plots (Knutson and Lioy, 1989). At EML we use a computer program, UNFOLD.PS, to construct smooth particle size spectra from the

impactor data. To do this, construct an impactor efficiency file from the curves given in Figure 7 of Knuth (1979b) per the instructions given in UNFOLD.PS, then follow the steps in Section 2.2.2.8, C6.

REFERENCES

Knuth, R. H.
"Comments on Inertial Impactor Calibration and Use"
in: *Aerosol Measurements,* University of Florida, Gainesville, pp. 108-116 (1979a)

Knuth, R. H.
"Calibration of a Modified Sierra Model 235 Slotted Cascade Impactor"
USDOE Report EML-360 (1979b)

Knutson, E. O. and P. J. Lioy
"Measurements and Presentation of Aerosol Size Distributions"
Lioy, P. J. and M. J. Y. Lioy (Editors)
in: *Air Sampling Instruments for Evaluation of Atmospheric Contaminants*
American Conference of Governmental Industrial Hygienists, Cincinnati, OH,
  pp. E1-E12, Seventh Edition (1989)

Marple, V. A. and K. Willeke
"Inertial Impactors"
D. A. Lundgren et al. (Editors)
in: *Aerosol Measurements*
University of Florida, Gainesville, pp. 90-107 (1979)

Marple, V. A., B. Y. H. Liu, and K. T. Whitby
"Fluid Mechanics of the Laminar Flow Aerosol Impactor"
J. Aerosol Sci., 5, 1-16 (1973)

Marple, V. A., B. Y. H. Liu, and K. T. Whitby
"On the Flow Fields of Inertial Impactors"
J. Fluids Eng., 96, 394-400 (1974)

2.2.2.5
MOUDI

A. Introduction.

Procedures are presented for using the micro-orifice uniform deposit impactor (MOUDI) in determining the particle size distribution of the decay products of radon and/or thoron gas. The MOUDI covers the size range of 50-5000 nm and complements the capabilities of the diffusion batteries (Section 2.2.2.6), which cover a range of 5-500 nm. To sample a broader size range, 0.5-5000 nm, the MOUDI may be used in parallel with the graded screen array, described in Section 2.2.2.7.

The MOUDI consists of two basic assemblies -- the cascade impactor itself, and its housing. The cascade impactor consists of an air inlet, eight impaction stages, and a backup filter, which is located in the base of the impactor. Each stage contains a removable impact plate for the stage above and a nozzle plate for the stage below.

Contained in the housing is a valve for controlling the flow through the impactor, two pressure gauges to monitor the flow, and a mechanism for rotating the impaction stages.

The table below gives the main characteristics of the MOUDI. Further information about the MOUDI can be found in Marple (1991).

CHARACTERISTICS OF THE MOUDI
(Manufacturer's data)

Stage	Cut Size, nm	No. of Nozzles	Pressure Reading
3A	3200	10	
4A	1800	20	
5A	1000	40	1250 Pa, upper gauge
6A	560	80	
7A	290	900	
8A	173	900	
B	97	2000	
BB	45	2000	45 kPa, lower gauge

C. Sampling procedure.

Preparation. Assemble the MOUDI for sampling by first inserting a filter in the backup filter holder at the base of the MOUDI, then by placing clean impaction plates on each stage starting from bottom to top of the impactor. Coating the plates with silicone spray (to prevent bounce) is necessary only in dry, dusty conditions. Finally, place the cover onto the upper stage.

Checking air flow rate. Start the pump and adjust the pressure reading to 1250 Pa for the upper gauge and 45 kPa for the lower one. If this cannot be accomplished, stop the pump and disassemble the MOUDI. Reassemble it after checking each stage to see if the gaskets are properly placed and greased. Then repeat the air flow test. If the pressure readings are satisfactory, proceed to sampling.

Caution: After testing the air flow rate, always check the backup filter, which is easily broken by a reverse airflow when the sampling pump is shut off.

Sampling. For radon progeny, the sampling time is usually 5 min, but 10 to

20 min is also used for low radon concentrations. For measurements of thoron progeny, a sampling period of 10 or more hours might be needed. In this case, use the rotating feature by turning on the switch at the base of the MOUDI. Use a stopwatch to time the sampling and keep the watch running to time the interval between sampling and counting.

Alpha counting. After sampling, disassemble the MOUDI starting with the upper stage and working downward to the base. Transfer the impaction plates into the alpha counters, being careful to keep them in order. Alpha count the plates simultaneously and analyze for activity using methods described in Procedure 2.2.4.6.

Generally, 10 alpha counters are needed. To complete sample transfer within 2 min, two experienced operators are needed.

Calculation of particle size. Follow the procedures given in Procedure 2.2.2.8, Sections C.6 or C.7. Use the calibration curves supplied by the manufacturer.

REFERENCES

Marple, V. A., Rubow, K. L. and Behm, S. M.
"A Microorifice Uniform Deposit Impactor (MOUDI): Description, Calibration, and Use"
Aerosol Science and Technology, <u>14</u>, 434-446 (1991)

2.2.2.6
DIFFUSION BATTERIES

A. Introduction.

We describe in this section EML's diffusion batteries (DB), and their use in measuring aerosol particle size distributions. There are five EML designs covering two main applications:

Type of diffusion battery	No. of channels or stages	Typical flow rate (L min^{-1})	Typical application
Multichannel disk-type	5	3	Radon progeny
Multistage disk-type	12	4	Aitken particles
Multichannel screen-type	5	25	Radon progeny
Multistage screen-type	11	4	Aitken particles
Multichannel carbon-type	5	283	Radon progeny

The multichannel designs, also called parallel diffusion batteries, are those in which air is drawn simultaneously through side-by-side samplers. Thus, all aerosol samples needed to construct the particle size distribution are collected simultaneously. In the multistage design, also called series, the aerosol samples are collected sequentially. The term Aitken particles refers to outdoor atmospheric particles with diameters < 0.2 µm. (However, diffusion batteries can be used for somewhat larger particles, either indoors or outdoors.) The term radon progeny particles refers to those few particles, typically 1 in 10^6, which carry short-lived radon progeny atoms. The batteries can also be used for other species, such as sulfate in airborne particles.

The type most commonly used in recent years is the multichannel screen battery, so this will be used for illustration in the discussion to follow. Consult Knutson and Sinclair (1979), Sinclair (1972), Sinclair and Hoopes (1975), and Sinclair et al. (1978) for further information on the other types.

We also use wire screens for a different but related radon progeny aerosol measurement (George, 1972).

B. Description of the multichannel screen diffusion battery.

Figure 2.1 shows the main components of the multichannel screen diffusion battery, and Figure 2.2 shows one of the components in the cross section. The system consists of five filter-type aerosol sampler units, each preceded by a different number of screens:

Sampler unit:	B0	B1	B2	B3	B4
Number of screens:	0	1	5	15	40

(Unit B0 is also called the reference filter.)

The screen used is a standard industrial twill-weave stainless-steel screen with the following dimensions:

Mesh	250 cm^{-1}
Wire diameter	$20 \, \mu\text{m}$
Screen thickness[*]	$50 \, \mu\text{m}$
Solid fraction	0.345
Diameter of flow area	10.16 cm

[*]As measured with a machinist's micrometer

C. Sampling procedure.

In normal use, the five samplers are loaded with clean $0.8 \, \mu\text{m}$ pore membrane filters (Type AA, Millipore Corp., Bedford, MA) and air is drawn at 25 L min^{-1} through each unit. As the air flows through the screens, particles are selectively removed from the airstream and deposited on the wires. Particles that escape collection by the screens are collected on the filters. For radon progeny the preferred sampling period is 5 min, but 10 or 20 min can be used if the concentration is low. For other species, the sampling period must be adjusted.

After sampling, the filters are removed and analyzed. When analyzing for radon progeny, any of the three alpha-counting protocols described in Section 2.2.4 can be used. As described there, it is important that the counting begin no more than 2 min after the end of sampling. This can be accomplished if two experienced operators are on hand at the time when the filters are transferred.

D. Calculations.

Recovery of the particle size distribution from the activity measured on the five filters is possible only with a quantitative knowledge of transport through the screens as a function of the particle size. We currently use the "CKK" equation (Cheng et al., 1980; Cheng and Yeh, 1983):

$$\Phi = \exp [-n \cdot m \cdot \ln (10)] \tag{1}$$

where

Φ = the fractional penetration through a stack of n screens

n = number of screens in the stack

m = $A_0 Pe^{-2/3} + A_1 R^2 + A_2 R^{2/3} Pe^{1/2}$

A_0 = 1.96, A_1 = 3.37, A_2 = 1.94

Pe = $u_f d_w/D$, the Peclet number

R = d_p/d_w, the interception parameter

D = $kTC/(3 \pi \mu d_p)$, the diffusion coefficient

k = 1.38×10^{-23} J $^{\circ}K^{-1}$, Boltzmann's constant

T = temperature in degrees absolute, normally $293^{\circ}K$

C = $1 + (\lambda/d_p)[2.514 + 0.800 \exp(-0.55 d_p/\lambda)]$, the Cunningham slip factor

d_p = particle diameter, m

d_w = wire diameter, m

λ = mean free path of air molecules, normally 66 x 10^{-9} m

μ = viscosity of air, normally 1.81 x 10^{-5} Pa sec

u_f = air approach velocity, m sec^{-1}.

The values for A_0, A_1, and A_2 apply to the standard (250 mesh cm^{-1}) screen - see Cheng and Yeh (1983) for other screens. The equation for the Cunningham factor is from Hinds (1982). In some of our computer programs we use an older equation with slightly different coefficients.

To analyze data from our diffusion batteries, we use one of the following two computer codes, which run on IBM-compatible personal computers:

Unfold.Pas - This is a Pascal program that uses the above-described Twomey algorithm. An alternative algorithm, the expectation-maximization algorithm, described by Maher and Laird (1985), is also available.

NMSimplx.Pas - This program, also in Pascal, makes use of the Nelder-Mead downhill simplex method to fit single or dual lognormal distributions to the data.

The logic involved in the first of these is the same as that in ExMaxDB.Pas, described in Knutson (1989).

REFERENCES

Cheng, Y. S. and H. C. Yeh
"Performance of a Screen-Type Diffusion Battery"
V. A. Marple and B. Y. H Liu (Editors)
in: *Aerosols in the Mining and Industrial Environment*
Ann Arbor Science Publishers, Ann Arbor MI (1983)

Cheng, Y. S., J. A. Keating, and G. M. Kanapilly
"Theory and Calibration of a Screen-Type Diffusion Battery"
J. Aerosol Sci., 11, 549-556 (1980)

George, A. C.
"Measurement of the Uncombined Fraction of Radon Daughters with Wire Screens"
Health Physics, 23, 390-392 (1972)

Hinds, W. C.
Aerosol Technology
John Wiley, NY (1982)

Knutson, E. O.
"Personal Computer Program for Use in Radon/Thoron Progeny Measurements"
USDOE Report EML-517, March (1989)

Knutson, E. O. and D. Sinclair
"Experience in Sampling Urban Aerosols with the Sinclair Diffusion Battery and Nucleus
  Counter"
Proceedings: Advances in Particle Sampling and Measurement, EPA Report No. EPA-
600/7-79-065, pp. 98-120, February (1979)

Sinclair, D.
"A Portable Diffusion Battery - Its Application to Measuring Aerosol Size
  Characteristics"
Am. Ind. Hyg. Assoc. J., 33, 729-735 (1972)

Sinclair, D. and G. S. Hoopes
"A Novel Form of Diffusion Battery"
Am. Ind. Hyg. Assoc. J., 36, 39-42 (1975)

Sinclair, D., A. C. George and E. O. Knutson
"Application of Diffusion Batteries to Measurement of Submicron Radioactive Aerosols"
in: *Airborne Radioactivity*
American Nuclear Society, La Grange Park, IL, Order No. 71001. pp. 103-114 (1978)

Figure 2.1. EML multichannel screen diffusion battery.

Aerosol Inlet (5.1 cm sec^{-1})

635 Mesh
Stainless Steel Screen
1, 5, 15 or 40
Layers

47 mm Filter

To Pump 25 L min^{-1}
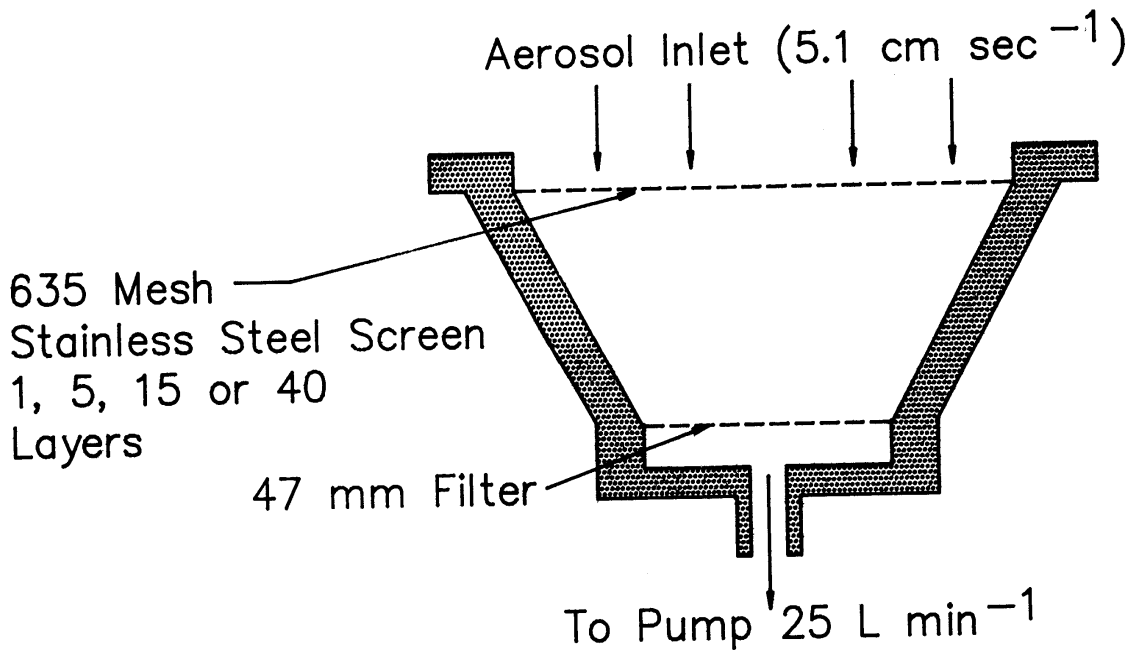
Figure 2.2. Cross section of the sampler units of the multichannel screen diffusion battery.

2.2.2.7
GRADED SCREEN ARRAY

A. Introduction.

The graded screen array (GSA, also called the graded screen diffusion battery) is used
for measuring the particle size of radon progeny in the range below 20 nm. The GSA is not
capable of dealing with larger sized aerosol particles, particularly those above 50 nm.
Therefore, the GSA should be used simultaneously with one of the diffusion batteries
shown in Section 2.2.2.6 or with the MOUDI impactor (see Section 2.2.2.5). For example,
the GSA used together with the MOUDI provides size information in the range of 0.5-5000
nm.

It is permissible to use the GSA alone, but only in laboratory studies in which radon
progeny or thoron progeny are sampled from a chamber free of larger aerosol particles.

B. Apparatus required.

1. One set of four circular stainless steel screens, each mounted on a metal ring — the
   properties of these screens are:

Screen label	Mesh, cm	Wire diameter, cm	Screen thickness, cm	Solid fraction
60	23.6	0.0187	0.0356	0.360
100	39.4	0.0108	0.0249	0.308
200	78.7	0.0052	0.0135	0.275
635	250	0.0020	0.0050	0.345

Two ring sizes are available: small and large. The dimensions and recommended uses are shown below.

Size of Ring	Outside diameter, cm	Inside diameter, cm	Ring thickness, cm	Flow area, cm^{-2}	Typical flow rate, $L\ min^{-1}$	For use at radon concentration, $Bq\ m^{-3}$
Small	2.67	1.91	0.25	2.86	3	>1000
Large	5.19	3.96	0.38	12.3	10	>200

2. A holder for the size of the screen selected.

3. A supply of Metricel DM-800 filters of the same size as the selected screens, and an open-faced holder.

4. A pair of rotameter-type flow meters, one for the GSA and one for the open-faced filter, and a suction pump.

5. Five drawer-type scintillation alpha counters of the type described in Section 2.2.4,Table 2.4.

6. A personal computer equipped with Keithley-Metrabyte CTM-05 (or equivalent) pulse-counting hardware and the software program ALPHALOG, as described in Section 2.2.4.6.

7. A stopwatch.

C. Procedure

1. Set up and check out the alpha-counting equipment, as described in Section 2.2.4.3B.

2. Calibrate both flow meters as described in Section 2.2.4.2A.

3. Using the alpha counters, check the background activity of the wire screens (the background should be no higher than 0.2 counts min^{-1}). This step may be omitted if it is known that the screens have not been used in the past 24 h.

4. Mount the four screens into the holder so that the air will flow through in the sequence 60-100-200-635; orient the mounting rings so that the flush side faces upstream.

5. Set up the filter for sampling, as described in Section 2.2.4.2B; set up the screen-holder in the same way.

6. Draw samples simultaneously through the filter and the screens, as described in Section 2.2.4.6.

7. After sampling, transfer the screens and the filter into separate alpha counters, and perform minute-by-minute counting using ALPHALOG, as described in Section 2.2.4.6.

8. Use program RWRENN6.EXE to calculate activity concentrations from the above count data.

9. Use this data to calculate particle size distributions, as described in Section 2.2.2.8. This method has been used to measure the diffusion coefficient of unattached radon progeny in filtered room air (George, 1994).

D. Quality Control.

1. For the air sampling and alpha counting steps, follow the quality control procedures outlined in Section 2.2.4.2A and 2.2.4.3B.

2. Whenever possible, take duplicate or triplicate samples from each experimental condition.

REFERENCES

George, A. C. and E. O. Knutson
"Particle Size of Unattached Radon Progeny in Filtered Room Air"
Radiation Protection Dosimetry, <u>56</u>, 119-121 (1994)

2.2.2.8

MATHEMATICAL ANALYSIS OF PARTICLE SIZE DATA

A. Introduction.

This section describes the steps needed to calculate particle size distributions from samples taken with the MOUDI impactor (Section 2.2.2.5), a diffusion battery (Section 2.2.2.6), and a graded screen array (Section 2.2.2.7), used individually or in combination. It can also be used for Sierra impactor data (Section 2.2.2.4).

The mathematical algorithms used in these calculation programs are described by Maher and Laird (1985), Solomon and Ren (1992), Knutson (1991), and Twomey (1975). Early versions of some of these programs are given in Knutson (1989). The full set of updated programs is available on a disk labeled EOKEMLUS.DOE.

B. Equipment and supplies needed.

1. An input data file (on disk) produced by the program RWRENN6.EXE.

2. Copies (on disk) of the files ABC^INP2.EXE, NMSIMPLX.EXE , UNFOLDPS and EMLMOUDI.EFF.

3. An IBM-compatible personal computer, preferably 386 or better.

C. Procedure.

1. Prepare the computer for this task as follows:

   a. From the root directory of the computer's C: drive, make a directory called UTIL and copy the above .EXE files into the UTIL directory.

   b. Include the UTIL directory in the PATH statement.

    c. From the root directory, make a directory called (for example) SIZEDATA and copy the above-mentioned input data file and the EMLMOUDI.EFF file into this directory.

    d. Change to the SIZEDATA directory.

2. Type ABC^INP2; when requested, supply:

-   the name of the RWRENN6 output file;
-   the number of data points in each data set (that is, the number of alpha counters that were used to collect the data);
-   whether or not the RWRENN6 program was instructed to analyze for thoron as well as radon progeny;
-   answer Y to the question about mapping.

This procedure produces a new file, with name ending in the character ^, that is ready for use by either NMsimplx or UnfoldPS.

From this point on, the procedure differs according to which sampler or combination of samples was used.

Sampler	Program to use	Option
Diffusion battery	NMsimplx	Classical RnP
Graded screen array	NMsimplx	Unimodal
Diffusion battery plus graded screen array	Nmsimplx	Bimodal (fixed, common geometric standard deviation, GSD)
MOUDI impactor	UnfoldPS	
MOUDI plus graded screen array	UnfoldPS	

   Both of these programs require that certain information be entered from the keyboard.
When answering a Y or N question it is not necessary to press ENTER. Other responses
require pressing the ENTER key to signal completion of the response. For many
questions, the proper response will be obvious; these are not discussed in the itemized
procedure below.

 3. <u>Diffusion Battery Alone</u>. As indicated above, use NMSIMPLX. Supply data as
    indicated below.   Type:

    -    a mask, such as *.??^, for the input data file,
    -    the number of the input data file, or 0 to try another mask,
    -    Y in answer to the question about error terms,
    -    A to select diffusion battery,
    -    a digit from 1 to 5 to specify which diffusion battery,
    -    the flow rate in L min^{-1},
    -    the uncertainty in the flow rate (normally about 3% of the flow rate itself),
    -    5 to select the Classical RnP,
    -    Y in answer to the question about default starting values,
    -    0 in answer to the question about Monte Carlo replications,
    -    or N, as appropriate, for the default temperature and pressure,
    -    Y in answer to each question about acceptable data quality.

The progress of the calculation can be viewed on the screen as NMsimplx runs. The results will also be stored in an ASCII file with an extension .NM5. The program produces an uncertainty estimate for each of the calculated parameters; this estimate is printed directly below the value of parameter.

4. <u>Graded Screen Array Alone</u>. Use NMsimplx. Type in the data indicated below.

- a mask, such as *.??^, for the input data file,
- the number of the input data file, or 0 to try another mask,
- Y in answer to the question about error terms,
- B to select graded screen array,
- N in answer to the questions about adding screens and showing rules,
- BCDE X in answer to prompt for GSconfig,[1]
- Y in answer to the question about alpha counting,
- the flow rates in L min^{-1},
- the uncertainty in the flow rate(s) (normally about 3% of the flow rate itself),
- N to the question about inlet losses,
- the flow area of the screens that were used,
- 1 to select unimodal,
- N in answer to the question about default starting values,
- 1 in answer to the request for a starting diameter,
- Y or N, as desired, to the question about switching to diffusion coefficient, (if no, answer Y or N, as appropriate, to the next question about temperature and pressure),
- 0 in answer to the question about Monte Carlo replications,
- Y in answer to each question about acceptable data quality.

The program will run and produce a file as described under Diffusion Battery Alone, except that the file extension will be .NM1.

---

[1] *This is the proper response if: the four screens were stacked into one holder during the sampling, such that the air flowed through in the sequence B-C-D-E; the filter was in a separate holder; and the data in the input file are in the sequence B,C,D,E,X Otherwise, answer Y to the question about screen rules, and use these rules to decide how to answer the GSconfig prompt.*

5. <u>Diffusion Battery plus Graded Screen Array</u>. Use NMsimplx. Type in the information itemized below.

- a mask, such as *.??^, for the input data file,
- the number of the input data file, or 0 to try another mask,
- Y in answer to the question about error terms,
- AB to select the combination of diffusion battery and graded screens,[1]
- a digit from 1 to 5 to specify which diffusion battery,
- the diffusion battery flow rate in L min^{-1},
- the uncertainty in the above flow rate (normally about 3% of the flow rate itself),
- N in answer to the questions about adding screens and showing rules,
- BCDE X in answer to prompt for GSconfig,[2]
- Y in answer to the question about alpha counting,
- the graded screen flow rate(s) in L min^{-1},
- the uncertainty in the above flow rate(s) (normally about 3% of the flow rate itself),
- the flow area of the screens that were used,
- 2 to select bimodal with fixed, common GSD,
- 1.5 in response to the prompt for a value of GSD,
- N in answer to the question about default starting values,
- 1 100 in answer to the request for a two starting diameters,
- 0 in answer to the question about Monte Carlo replications,
- Y or N, as appropriate, to the question about temperature and pressure,
- Y in answer to each question about acceptable data quality.

The output file from the above steps will have the extension .NM2.

---

[1] *This is the proper response if the diffusion battery data <u>precedes</u> the graded screen data in the input data file. Otherwise interchange A and B.*

[2] *This is the proper response if: the four screens were stacked into one holder during the sampling, such that the air flowed through in the sequence B-C-D-E; the filter was in a separate holder; and the data in the input file are in the sequence B,C,D,E,X. Otherwise, answer Y to the question about screen rules, and use these rules to decide how to answer the Gsconfig prompt.*

6. <u>MOUDI, alone or with a reference filter</u>. Use UnfoldPS. Before proceeding, review the input data file and make note of which impactor stages are represented in the file. (Often the upper stages are not alpha-counted.) Also review the EMLMOUDI.EFF file and make note of the 50% cut points of the first and last stage for which there is data in the input file. Type in the following information:

- a mask, such as *.??^, for the input data file,
- the number of the input data file, or 0 to try another mask,
- Y in answer to the question about error terms,
- C to select impactor,
- 40 in answer to the question about smallest size,[1]
- 4000 in answer to the question about largest size,[2]
- 2 to select lognormal basis functions,
- 1.3 in answer to the question about GSD,
- Y or N, as appropriate, concerning the temperature and pressure,
- EMLMOUDI.EFF in response to the prompt for an info file,
- 1 in answer to the question about particle density,[3]
- Y or N, as appropriate, to the questions about data present in the input file,
- Y in answer to the questions about data quality,
- 0 for the number of Twomey iterations,
- 100 for the number of E-M iterations,
- 0.00005 for the convergence criterion,
- N in answer to the question about skipping negative values,
- 0 in response to the prompt about Monte Carlo error study,
- Y in answer to the question about long-form output,
- ENTER in answer to the request for file name,
- Y in answer to the question about short-form output,
- ENTER in answer to the request for file name,
- Y in response to the question about 50-line screen.

---

[1] *This value should be no smaller than 80% of the 50% cut-point of the last impactor stage for which there is data in the input file.*

[2] *This value should not exceed 120% of the 50% cut-point of the first impactor stage for which there is data in the input file.*

[3] *Entering 1 will cause the results to be presented in terms of equivalent unit-density spheres, i.e., the aerodynamic equivalent diameter. Entering the actual density of the particle will cause the results to be presented in terms of the Stokes equivalent diameter.*

As UnfoldPS runs, the progress of the current calculation is shown near the bottom of the screen. The upper part of the screen shows the results of the previous calculation. When finished, the long-form output can be found in a file with extension ending in &, and the short-form output will be in a file with extension ending in.

7. <u>MOUDI plus Graded Screen Array</u>. Use UnfoldPS. Before proceeding, review the input data file and the EMLMOUDI.EFF file as described in the last section. Then type in the following information -

- a mask, such as *.??^, for the input data file,
- the number of the input data file, or 0 to try another mask,
- Y in answer to the question about error terms,
- CB to specify the type of sampler,[1]
- 0.5 in answer to the question about smallest size,
- 4000 in answer to the question about largest size,[2]
- 2 to select lognormal basis functions,
- 1.3 in answer to the question about GSD,
- Y or N, as appropriate, concerning the temperature and pressure,
- EMLMOUDI.EFF in response to the prompt for an info file,
- 1.5 in answer to the question about particle density,[3]
- Y or N, as appropriate, to the questions about data present in the input file,
- N in answer to the questions about adding screens and showing rules,
- BC in answer to the prompt for GSconfig,[4]
- Y in answer to the question about alpha counting,
- the graded screen flow rate in L min^{-1},

---

[1] *This is the correct response if the impactor data precedes the grades screen data in the input file; otherwise, interchange B and C.*

[2] *This value should not exceed 120% of the 50% cut-point of the first impactor stage for which there is data in the input file.*

[3] *This value of particle density is approximately correct for indoor aerosols; the results will be g iven in terms of Stokes equivalent diameter, which is appropriate when combining data from impactors and diffusion batteries, or graded screens.*

[4] *This assumes that data are available only from screens B and C, and that these two screens were stacked during sampling, such that the air flowed through in the sequence B-C. Otherwise, read the "screen rules" to determine the proper entry for GSconfig.*

- the uncertainty in the above flow rate (normally about 3% of the flow rate itself),
- the flow area of the screens that were used,
- Y in answer to the questions about data quality,
- 0 for the number of Twomey iterations,
- 1000 for the number of E-M iterations,
- 0.00005 for the convergence criterion,
- N in answer to the question about skipping negative values,
- 0 in response to the prompt about Monte Carlo error study,
- Y in answer to the question about long-form output,
- ENTER in answer to the request for file name,
- Y in answer to the question about short-form output,
- ENTER in answer to the request for file name,
- Y in response to the question about 50-line screen.

D. Quality Control.

1. Using a text-viewing or text-editing program, examine the output file from NMsimplx or from UnfoldPS. Starting at the end of the file, spot-check the column titled "RawData" to ensure that the input data have been properly matched to the stages of the sampling device. It will be necessary to consult the RWeENN6 output file to determine the proper matching.

2. Examine each block of results in the output file. Any block of data that has more than one negative input datum should be discarded. Examine also the "ChiSqr" value, which is a measure of the internal consistency of the data. If this number is > 25, the data block should be discarded unless the reason for the high value can be identified and fixed.

3. Similarly if the ChiSqr value is < 1, the data block should be discarded because the uncertainties in the input data are too large to support calculation of the particle size distribution.

REFERENCES

Knutson, E. O.
"Personal Computer Programs for Use in Radon/Thoron Progeny Measurements"
DOE Report EML-517 (1989)

Knutson, E. O.
"Application of the Expectation-Maximization Algorithm to the Processing of Cascade
  Impactor Data: The Method of Lognormal Components"
J. Aerosol Sci., 22, 267-270 (1991)

Maher, E. F. and N. M. Laird
"Algorithm for Reconstruction of Particle Size Distributions from Diffusion Battery
  Data"
J. Aerosol Sci., 16, 557-570 (1985)

Solomon, S. B. and T. Ren
"Counting Efficiencies for Alpha Particles Emitted from Wire Screens"
Aerosol Sci. and Tech., 17, 69-83 (1992)

Twomey, S.
"A Comparison of Constrained Linear Inversion and a Iterative Nonlinear Algorithm
  Applied to the Indirect Estimation of Particle Size Distributions"
J. Comput. Phys., 18, 188-200 (1975)

2.2.3 RADON AND THORON

2.2.3.1
INTRODUCTION

The procedures and instruments commonly used at EML for measuring radon are presented here. The main sampling methods are grab, two filter tube, continuous, and integrating. Both active and passive methods are included. These methods are used in laboratory applications as well as in situations requiring mobility and portability.

2.2.3.2
GRAB SAMPLING FOR RADON

Three sizes of scintillation flasks, all based on a design described in George (1976), are used at EML. Typical characteristics are shown in Table 2.1. The flasks are made at EML from methylmethacrylate stock. The bottom plate, made from sheet, is cemented to the body, made from tubing. The top plate, also from sheet, slides into the body and is sealed with an O-ring. The top plate is fitted with two petcock valves. All three flasks are designed to be counted on 12.7-cm phototubes.

As the radon inside the flask decays, most of the radon progeny that are produced plate out on the interior surfaces of the flask. The alpha particles from radon and radon progeny interact with the ZnS(Ag) atoms to produce scintillations which are detected, amplified, and converted to an electrical signal by the phototube.

A. Flask coating procedure.

In this procedure, we place a thin phosphor coat on all internal surfaces. The thin coating on the bottom plate adds 15-20% to the sensitivity. The materials and steps used in this process are listed below.

Coating materials*:*

1. Silver activated zinc sulfide phosphor (Wm. B. Johnson Associates, PO Box 472, 216 Edgar Ave., Ronceverte, WV 24970).

2. Bonding solution (Caution-Flammable-Use Hood). Dissolve 30 mL of silicone fluid (Dow Corning, Midland, MI; Silicone Fluid 200) in 285 mL of reagent benzene plus 285 mL of reagent cyclohexane. Stir until the silicone is completely dissolved. This is enough to coat about 100 flasks.

Coating steps (use a hood):

1. Clean the flask with a nonabrasive cleaner. [A plastic cleaning foam (REN RP-70 cleaner) has been found to be excellent.]

2. Remove cover, add 50 mL of bonding solution, replace cover, close both valves, and rotate the flask slowly until all surfaces including the bottom are coated. Open valves and remove cover.

3. Pour out excess bonding mixture into a second clean flask or return to the stock bottle. Let flask air dry for a few minutes.

4. Add 15-20 g of phosphor, replace cover, and close valves.

5. Shake gently until all surfaces are coated. Open valves and remove cover.

6. Pour out the loose powder and blow off excess phosphor from the flask and from the cover with clean air.

7. Replace cover and purge the flask with aged (radon free) air or nitrogen for a few minutes. Close the valves and store the flask for use.

8. When background becomes excessive, wipe out phosphor with clean tissue, clean as above, and rephosphor.

B. Sampling and counting procedures.

1. Connect a high collection efficiency filter to one valve of the flask to remove particulates and radon progeny. Connect the other valve to an air pump and draw air at 2-10 L min^{-1} for 2-5 min for scintillation flasks of 165-2000 mL.

2. Turn off pump and close both valves. An alternate method for filling the flask is to evacuate it down to 133 Pa (1 mm Hg) of pressure prior to filling with the test atmosphere.

3. Count the scintillation flask on a 12 cm phototube beginning 3 h after sampling.

4. To convert the counting rate to radon concentration, use the calibration factor (C_f) obtained from a test in a known radon environment and the appropriate decay-correction. Each scintillation flask should have its own C_f which can be influenced by its size and by the phototube assembly. It is very important that the entire system (scintillation flask, and phototube) is calibrated as a unit. Typical C_f for the three scintillation flasks used at EML are shown in Table 2.1.

5. Determine the radon concentration at the time of sampling by applying the decay correction (D_f):

$$D_f \; = \; \exp\left(\frac{-0.693\,t}{T_{1/2} \;\; \text{for radon}} \right) \; = \; \exp\left(-0.0075\,t\right) \tag{1}$$

where t is the time in hours from the midpoint of sampling to the midpoint of counting, and $T_{1/2}$ is the half-life of radon (91.7 h).

REFERENCE

George, A. C.
"Scintillation Flasks for the Determination of Low-Level Concentrations of Radon"
Proceedings of Ninth Midyear Health Physics Symposium, Denver, Colorado, February (1976)

2.2.3.3
TWO-FILTER TUBE SAMPLING FOR RADON AND THORON

The two-filter tube sampling method described in this section is used for simultaneous measurements of airborne radon and thoron (Knutson et al., 1994).

A. Description.

The sampling tube (9.7 cm diameter x 61 cm in length) has two filter heads. The front end of the tube is fitted with a 10.5-cm filter holder to accommodate two 10.5-cm high efficiency glass fiber filters mounted in series for the collection of all particulates including radon and thoron progeny. The back end of the tube is fitted with a 4.7-cm filter head assembly to hold a 4.7-cm diameter high efficiency Metricel membrane filter paper. This filter has very low alpha-radioactivity background (0.05 counts min^{-1}) and antistatic electrical properties. The background counting rate of the back-end filter is critical in measuring the expected low counting rate from the buildup of radon and thoron progeny originating inside the tube. The sampling flow rate, ranging from 30-50 L min^{-1}, is high enough to minimize the loss of ^{216}Po and ^{218}Po atoms to the wall of the tube and to maximize the collection on the back-end filter.

B. Sampling and counting.

1. Mount two 10.5-cm diameter glass fiber filters in series in the front end of the tube and a single 4.7-cm Metricel membrane filter in the back end of the tube.

2. Connect the intake of an air pump to the back end of the tube and sample for 5-6 h.

3. At the end of sampling, transfer the back-end filter into a 5-cm diameter gross alpha scintillation counter and begin to count as soon as possible in short time intervals (5-10 min) for 4-6 h. One convenient way to do this is to make use of a portable personal computer equipped with pulse-counting hardware and the software program ALPHALOG (see Section 2.2.4.6). The sooner the counting begins the better the precision that is obtained for radon measurements; for thoron, the sensitivity increases with counting times of several hours.

C. Calculations.

The concentrations of radon and thoron are calculated from the radioactivity measured on the back-end filter using a computer program JWTI, which is based on the recursion formula method of Samuelson (1987). This method is very flexible employing different sampling regimes and counting intervals. The calculated penetration (F_f) of ^{216}Po from thoron and ^{218}Po from radon is governed by the flow rate and the value for the diffusion coefficient, D, used for these isotopes. The relationship between D and F_f is obtained from Table 2.2, using the value of the parameter μ given by the expression

$$\mu \;=\; \frac{\pi \times D \times L}{Q}$$

where

$\pi \;=\; 3.14.$

$D \;=\;$ diffusion coefficient, $cm^2\ sec^{-1}$ ($0.085\ cm^2\ sec^{-1}$)

$L \;=\;$ length of tube, cm (61 cm)

$Q \;=\;$ sampling flow rate, $cm^3\ sec^{-1}$ ($500\text{-}800\ cm^3\ sec^{-1}$)

The value of $D = 0.085\ cm^2\ sec^{-1}$ is used for very fresh radon progeny. The concentration of radon or thoron is underestimated by 3%, 4.5%, and 6.0% if the value used for $D = 0.06$, 0.05, and $0.043\ cm^2\ sec^{-1}$, respectively.

A computer program JWT1 is used to calculate both radon and thoron concentrations in $Bq\ m^{-3}$. The lower limit of detection is 5 $Bq\ m^{-3}$ for both radon and thoron. For concentrations higher than 200 $Bq\ m^{-3}$, a smaller two-filter tube can be used (Thomas, 1970).

REFERENCES

Knutson, E. O., A. C. George, P. Shebell and C. V. Gogolak
"EML Thoron Gas Measurements"
Radiation Protection Dosimetry, <u>56</u>, 263-266 (1994)

Samuelson, C.
"Applications of a Recursion Formula to Air Sampling of Radon Daughters"
Nuclear Instruments and Methods in Physics Research, <u>A262</u>, 457 (1987)

Thomas, J. W. and P. C. LeClare
"A Study of the Two-Filter Method for Radon-222"
Health Phys., <u>18</u>, 113 (1970)

2.2.3.4
CONTINUOUS RADON MONITORING

Continuous reading radon monitors are ideal for measuring varying concentrations of radon over a long period of time. The scintillation flask used for radon grab sampling can be adapted to measure radon continuously by sampling at flow rates of 1-2 L min^{-1} as described in Thomas and Countess (1979). Any of the three flasks used at EML can serve this purpose. The flask is mounted on top of the phototube in light-proof housing. Sample air is drawn continuously through the flask by means of a pump, with a high efficiency filter mounted inline to remove dust particles and radon progeny. The number of alpha counts accumulated at the end of each counting interval, usually 30-60 min, is stored in a computer or printed out on paper tape.

The entire system is calibrated in a continuous operation mode to obtain the appropriate C_f at a fixed flow rate and at varying radon concentrations. In an atmosphere in which the concentration of radon changes drastically, the calibration of the whole system becomes tedious. However, with the aid of modern desk computers the average radon concentration during the sampling interval can be calculated accurately. This is accomplished by means of an equation that takes into consideration the number of counts obtained in preceding sampling intervals (Thomas and Countess, 1979).

The C_f from four continuous 2-L scintillation flask radon monitors used in the EML radon calibration facility at flow rates of 1 L min^{-1} range from 0.0019-0.0020 counts min^{-1} per Bq m^{-3} (4.2-4.5 counts min^{-1} per pCi L^{-1}). The range in factors results from differences in phototubes and scintillation flasks.

REFERENCE

Thomas, J. W. and R. J. Countess
"Continuous Radon Monitor"
Health Phys., 36, 734 (1979)

2.2.3.5
INTEGRATING RADON MONITORS

The activated carbon monitor is used to obtain the average radon concentration over a 1-3 day period of passive sampling. The original design (George, 1984) was based on the M11 charcoal canister. Although there are currently many variants of the activated carbon monitor, the device described below is the one most used at EML.

A. Description.

This monitor consists of the following materials: (1) a metal can with a lid (1.2-cm diameter by 2.9-cm deep); (2) 75 g of 8-16 mesh activated carbon (Calgon, Pittsburgh, PA); (3) metal screen (80 mesh) to keep the carbon in place; (4) removable internally expanding retaining ring to secure the screen; and (5) a strip (35 cm) of vinyl tape for sealing the lid to the can when closed. After loading and after each use, the carbon monitor is heated at $100^{\circ}C$ for a minimum of 10 h to drive off any previously adsorbed radon and water.

The counting system consists of: (1) an 8.0-x-8.0 cm NaI(Tl) detector inside a 9-cm lead shield coupled to a compact multichannel pulse-height analyzer and a printer. The counting system is operated with the discriminator set to include gamma energies between 0.23 and 0.72 MeV, spanning the peaks of ^{214}Pb (0.242, 0.294, and 0.352 MeV) and ^{214}Bi (0.609) MeV. The normal counting period is 10 min.

To determine the counting efficiency of the NaI detection system, a monitoring device is sealed after spiking with a known quantity of radon or ^{226}Ra. This becomes the standard source with the proper counting geometry. A different standard source will be required

for any change in the configuration of the monitor. Typically, the efficiency of the EML canister and counter combination is 0.117 counts per radon atom decay.

B. Calibration.

The monitors must be calibrated because the response depends on the length of exposure, relative humidity during the exposure, and on the type of carbon used. (Temperature effects encountered indoors are, however, insignificant because of the limited temperature range encountered.) Calibration is performed for several relative humidities from 20-90% and for 1-3 days of exposure. This calibration is required whenever new samplers are built using a new batch of activated carbon.

Each combination of exposure time and humidity yields a C_f calculated from the following equation:

$$C_f = \frac{1000\,N}{60\,E\,Rn\,D_f}$$

where

$1000 = $ L m^{-3},

$N = $ net counting rate (counts min^{-1}),

$60 = $ sec min^{-1},

$E = $ NaI detector efficiency, typically 0.117 counts sec^{-1} Bq^{-1} Rn (0.26 counts min^{-1} pCi^{-1}), and

$Rn = $ radon concentration in Bq m^{-3},

$D_f = $ decay factor from the midpoint of exposure to the midpoint of counting (see Equation 1, Section 2.2.3.2).

Typical results are shown in Table 2.3. The amount of water adsorbed, shown in the left column, was determined from the difference in weights before and after exposure.

C. Use of device.

Before exposure the monitoring device is heated to regenerate it. Exposure times longer than 3 days should be avoided because water competes with radon adsorption and the effectiveness of the device as an integrating monitor will diminish.

To calculate the concentration of radon from the measured count rate and measured water uptake, Equation 2 is applied after solving for radon:with the symbols as

$$Rn = \frac{1000\,N}{60\,E\,C_f\,D_f}$$

defined before. Use Table 2.3 to obtain the C_f. The 1 σ counting error is calculated from the following expression:

$$\sigma = \frac{(Gross\ Counts\ +\ Background\ Counts)^{1/2}}{Gross\ Counts\ -\ Background\ Counts} \times 100\%$$

When exposed for 3 days and counted 3 days later, the device described here has a lower limit of detection of 7.4 Bq m^{-3}. Other versions of the device that contain better adsorptive carbon can measure radon concentration levels at 3.7 Bq m^{-3} or less.

REFERENCE

George, A. C.
"Passive Integrated Measurement of Indoor Radon Using Activated Carbon"
Health Phys., 46, 867 (1984)

2.2.3.6
QUALITY CONTROL

To verify the measurement accuracy of all monitoring devices for radon and thoron, periodic intercomparison measurements are made with pulse ionization chambers (Fisenne and Keller, 1985). The pulse ionization chambers are the primary calibration instruments traceable directly to the NIST through use of their standard ^{226}Ra reference solutions. The accuracy of the thoron concentrations is obtained through intercomparisons (Knutson et al., 1994).

REFERENCES

Fisenne, I. M. and H. W. Keller
"The EML Pulse Ionization Chamber System for Radon-222 Measurements"
USDOE Report EML-437, March (1985)

Knutson, E. O., A. C. George, P. Shebell and C. V. Gogolak
"EML Thoron Gas Measurements"
Radiation Protection Dosimetry, 56, 263-266 (1994)

TABLE 2.1

TYPICAL CHARACTERISTICS OF THE THREE SIZES OF EML
SCINTILLATION FLASKS

Flask volume (L)	0.165	0.415	2.00
C_f:			
counts min^{-1} Bq m^{-3}	0.022	0.054	0.135
counts min^{-1} pCi L^{-1}	0.8	2.0	5.0
LLD (Bq m^{-3})	7.4	3.7	3.0

TABLE 2.2

VALUES OF F_f AS A FUNCTION OF $\mu = \pi \times D \times L/Q$

μ	F_f	μ	F_f
0.005	0.877	0.25	0.420
0.008	0.849	0.30	0.384
0.010	0.834	0.35	0.349
0.020	0.778	0.40	0.324
0.030	0.737	0.45	0.302
0.040	0.705	0.50	0.282
0.050	0.678	0.60	0.248
0.060	0.654	0.70	0.220
0.070	0.633	0.80	0.220
0.080	0.614	0.90	0.178
0.090	0.596	1.00	0.162
0.100	0.580	1.50	0.110
0.120	0.551	2.00	0.083
0.140	0.525	2.50	0.067
0.160	0.502	3.00	0.056
0.180	0.481	4.00	0.042
0.200	0.462	5.00	0.033

TABLE 2.3

C_f: EFFECTIVE VOLUME SAMPLED AS A FUNCTION OF EXPOSURE
TIME AND WATER ADSORBED

Water adsorbed (g)	Effective sample volume (L) for various exposure times		
	1 day	2 days	3 days
0.500	144	216	259
0.700	143	213	255
1.000	142	210	251
1.500	140	204	246
1.700	138	202	242
2.000	137	199	238
2.500	135	196	233
3.000	132	190	229
3.500	131	187	225
4.000	130	184	220
4.500	127	181	216
5.000	125	179	207
6.000	121	167	199
7.000	118	161	190
8.000	114	153	181
9.000	109	144	173
10.000	105	135	164
11.000	101	127	156
12.000	096	121	151

2.2.3.7
^{220}Rn AND ^{222}Rn IN SOIL GAS

APPLICATION

This section describes the procedures used to determine soil gas ^{220}Rn and ^{222}Rn concentrations using commercially-available scintillation cells and photomultiplier tube apparatus (referred to as "^{222}Rn detector" in the following procedure).

A. Apparatus.

1. Scintillation cells (Scintrex, Ltd., 222-T Snidercroft Rd., Concord, ON, Canada, nee EDA, Model #RDX-013, or equivalent).

2. Photomultiplier tube and counting apparatus (Scintrex, Ltd., 222-T Snidercroft Rd., Concord, ON, Canada, Model #RDA-200, or equivalent).

3. Stainless-steel sampling tubes, 0.95 cm ID, various lengths.

4. Portable battery-operated pump (Du Pont E. I. De Nemours and Co., Route 87 S., PO Drawer Z, Fayetteville, NC 28302, Model #2500B, or equivalent) capable of ~ 2.0 L min^{-1} at Δp of 10 kPa.

5. Flow meter, 0-5 L min^{-1} (Cole-Parmer, or equivalent).

6. Pressure gauge, 0-25 kPa (Dwyer Instruments, Inc., PO Box 373-T, Michigan City, IN 46360, or equivalent).

7. Short section of Tygon tubing (or equivalent) filled with dessicant held in place by glass wool, fitted at one end with an in-line filter holder containing Millipore (or equivalent) 0.8 µm filter to remove progeny from initially entering scintillation cell.

8. Computer program RNTNCAL.FOR (Hutter, unpublished) or equivalent, for calculating ^{220}Rn and ^{222}Rn concentrations from scintillation cell counts.

B. Procedure.

1. Emplace sampling tubes into the soil using reamer to pound, ensuring no soil is retained in tube, or alternatively, drill hole using auger and insert tube, tamping soil to minimize cavities along tube walls.

2. Obtain a 5-min background count for each scintillation cell to be used before the sample is drawn at the measurement site.

3. Obtain the soil gas sample using a portable battery-operated pump, according to the apparatus arrangement shown in Figure 2.3, with a flow rate of between 1 and 3 L min^{-1} for 1 min. Measure and record the flow rate and the volume of the sample tubing. The sample is drawn directly into scintillation cells that have been previously inserted into the ^{222}Rn detector.

4. Turn the pump off, remove the tubes to the scintillation cell and screw on the cap to the ^{222}Rn detector.

5. Obtain a 1-min count (see Notes 1 and 2).

6. Remove the scintillation cell from the ^{222}Rn detector, and obtain another sample using a different scintillation cell, or see Note 3 to reuse a cell.

7. Obtain a 5- or 10-min count at least 5 min after the soil gas sample was drawn into the scintillation cell, and no longer than 3 or 4 days. Typically, these counts are obtained either on-site at the end of collecting all samples, or within an hour to two at a convenient (i.e., warm and dry) site.

8. Calculate the concentration of ^{220}Rn and ^{222}Rn, correcting for decay of ^{220}Rn during sampling (Hutter, 1995). This requires knowledge of the flowrate and sample tube volume. A computer program, RNTHCALC.FOR, is available to do this calculation.

Notes:

1. The time between the end of pumping and the start of counting must be as short as possible (< 90 s; 1 1/2 half-lives of ^{220}Rn). Typically, this delay time is about 10 sec. Record the time between the end of pumping and the start of counting to within ± 1 s.

2. The counts min^{-1} from the sample must turn out to be at least 3 times the background. Obtain a new sample if this requirement is not met, either using a different scintillation cell, or by waiting until the background decreases.

3. If the 5-min count is obtained 5 min after sampling is ended, the cell can then be purged with outdoor air and a new background count obtained, to be reused for the next sample, provided that the new background check shows the sample counts min^{-1} to be at least 3 times the background.

C. QA/QC.

Duplicate samples, defined as measurements performed on the same sampling hole on the same day ~ 10 to 15 min apart using a different scintillation cell, are performed at a rate of 1 in 10 for the purpose of assessing measurement precision. Second countings of samples are performed at approximately the same rate in order to ensure cell integrity with regard to leakage and to assess statistical counting uncertainties.

Calibrations of scintillation cells/^{222}Rn detector apparatus are performed on at least a semiannual basis using the EML Radon Gas and Radon/Thoron Progeny Facilities for Testing and Research (see Section 6.2) or other radon chambers. At this time, it is useful to check for leaks in the scintillation cells by testing for consistency in the count rate, and by taking into consideration decay over several days. Approximately 1 in 5 cells obtained from a commercial vendor have been found to leak. Nearly all leaks can be sealed using epoxy around the window and all valve connections.

REFERENCES

Hutter, A. R.

"A Method for Determining Soil Gas ^{220}Rn (Thoron) Concentrations"

Health Physics, 68 835-839 (1995)

Hutter, A. R.

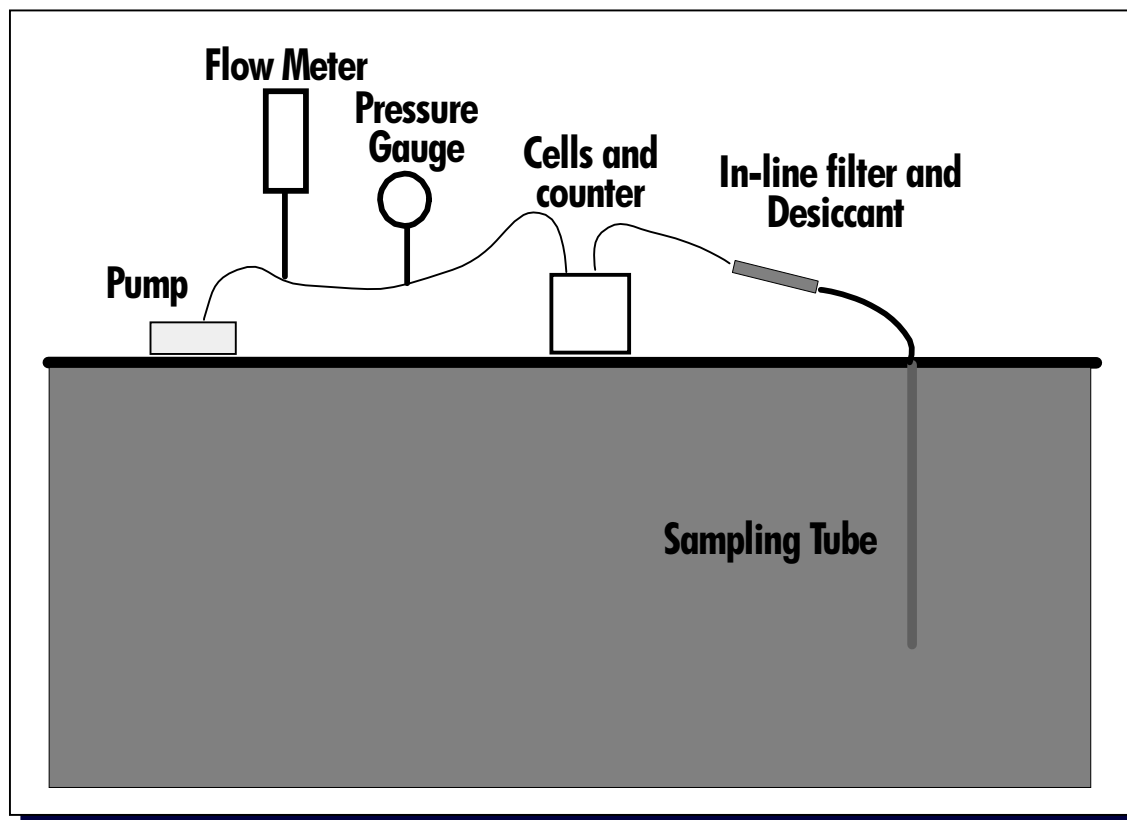"^{220}Rn/^{222}Rn Ratios as Indicators of Soil Gas Transport"

(unpublished data)

Figure 2.3. Schematic of soil gas sampling arrangement.

2.2.4 **RADON AND THORON PROGENY**

2.2.4.1
INTRODUCTION

Presented here are the three methods commonly used at EML for determining the concentration of radon and thoron progeny in air. All three methods - modified Kusnetz, modified Tsivoglou, and Raabe-Wrenn least squares - are based on collection in a known volume of air through a filter in a known time period, and on counting the radioactivity on the filter after sampling. The methods described are suitable for use in the laboratory, in homes, and in the workplace. The same sampling and counting equipment are used, but with different counting and calculational procedures.

Units: when specifying the concentration of individual progeny nuclides, the Systeme Internationale (SI) unit is Bq m^{-3}; when specifying the potential alpha energy concentration (PAEC) of radon or thoron progeny, the SI unit used is J m^{-3}.

These procedures have also been adopted by the World Health Organization (George, 1993).

2.2.4.2
SAMPLING TRAIN

All three methods require a sampling train such as that shown in Figure 2.4.

A. Air pump and flowmeter.

A portable air pump capable of moving 10-20 L min^{-1} through a high efficiency filter is required. The pump should be rugged, light weight, quiet, and operate from the voltage line in the building. A calibrated flowmeter to measure the flow rate through the filter holder assembly should be attached upstream of the intake of the pump. Calibration of the flowmeter is air density dependent and care should be taken if used at different altitudes. The pump flowmeter train assembly should be calibrated at the same elevation,

and at the same resistance that is caused by the filter and by the other component parts in the sampling train as shown in Figure 2.4. Never measure the air flow rate from the exhaust of the pump because of possible leakage in the pump housing.

B. Air filters and filter holder.

Use membrane filters with nominal pore size < 0.8 µm, or glass fiber with a collection efficiency of $> 99.9\%$. Membrane filters exhibit negligible alpha-particle absorption during alpha counting. Glass fiber filters allow deeper penetration of particles, but with the recommended flow rates, face velocity (< 100 cm sec^{-1}), and short sampling period, particle burial in the filter is negligible. Any other type of filter medium should be investigated for good surface deposition characteristics. In general, small pore filters exhibit the best surface collection, but also present the highest resistance to air flow. To compensate for this, use a higher capacity pump or a larger diameter filter and filter holder. The diameter of the filter can either be 2.5 or 4.7 cm depending on the size of the filter holder and the detector counter used. The connecting tubing between pump and filter holder should be a thick-walled pressure type hose. Filter holders should be of the open faced type to prevent the loss of radon progeny particles due to plate out upstream from the filter. The biggest error in measuring radon progeny by collection on a filter is often from a defective filter holder. To prevent this, make sure the filter holder-filter assembly is airtight and has the proper O-ring seal.

C. Timing device.

Sample collection timing is very important in the short sampling periods used for short-lived radionuclides. Errors may be significant if operators rely on their wristwatch and on their memory.

2.2.4.3
ALPHA PARTICLE COUNTING SYSTEM

A. Description.

Table 2.4 shows the main characteristics of the two alpha-particle counters most used at EML to measure the alpha activity deposited on a filter. Both counters are designed for gross alpha (total alpha) counting, in which the alpha particles from ^{218}Po (6 MeV) and ^{214}Po (7.7 MeV) are counted without discrimination.

Both counting systems consist of a simple scintillation disc (ZnS silver activated screen) mounted on or close to the face of a photomultiplier tube, and placed in close proximity to the filter. [In the TH-29-B (see Table 2.4), the scintillator is a separate Mylar disk coated on one side with ZnS; be sure the coated side faces the filter.] The photo-multiplier tube and preamplifier are housed in light-tight housing and are connected to a stable high voltage supply and a scaler.

B. Calibration.

Table 2.5 shows the characteristics of the certified alpha standard source most used at EML to periodically measure the efficiency of the counting systems. The counting efficiency is the ratio of the counting rate (in counts sec^{-1}) of the instrument to the known activity (in Bq) of the standard source. The EML counters described above have an efficiency of about 48%, but this figure should always be remeasured after the counters are moved to a field site.

Checking calibration is also a good quality assurance step. Measured efficiencies which are outside the range 47-49%, or which are not reproducible, are an indication of equipment malfunction and these counters should not be used.

Because the counters are energy-independent, efficiencies determined with the ^{241}Am or ^{239}Pu sources (alpha energies 5.53 MeV and 5.15 MeV, respectively) are believed to apply to radon/thoron progeny as well. For very precise work, the efficiency as measured above should be multiplied by the factor shown in Table 2.5 to correct for backscatter.

This correction takes into account the fact that the alpha emission from the front face of a metal plate is slightly higher than from the same activity deposited on a filter.

2.2.4.4
MODIFIED KUSNETZ METHOD

The simplest method is the Modified Kusnetz, in which a 5 min air sample is taken on a high efficiency filter to be counted for 5-10 min any time between 35-95 min after the end of sampling. This method differs from the original method (Kusnetz, 1956) in that a scaler is used in place of a ratemeter and the counting time is longer.

A. Procedure.

Before sampling and counting a sample, the operator should ensure that the entire sampling train is in good working order and that the counting system is calibrated and running properly. The counting efficiency of the system and its background counting rate should be determined as discussed earlier.

Using a 2.5 or 4.7 cm open-faced filter holder, mount a membrane or glass fiber filter. Attach the filter holder upstream of a calibrated flowmeter with the proper hose and connect to the intake of an air pump.

Sample air from the environment to be tested for 5 min at a rate of 10-20 L min^{-1}. After sampling, remove the filter containing the radon progeny from the filter holder and transfer it to the counting system for analysis. There is no urgency to do this since the operator has 35 min to commence counting. During that waiting period he/she may wish to take another sample in another part of the building using a new filter and the same filter holder and sampling equipment.

The modified Kusnetz method requires a single count of the filtered sample usually for 10 min in the counting interval from 35-45 min after the end of sampling, or at any other 10-min intervals between 35-85 min after the end of sampling.

B. Calculation.

The potential alpha energy concentrations (PAEC) can be calculated from the equation:

$$\text{PAEC in nJ m}^{-3} = N/(E \times T_s \times Q \times T_f) \qquad (1)$$

where

$$N \quad = \quad \text{net counts min}^{-1},$$

$$E \quad = \quad \text{fractional counter efficiency (counts sec}^{-1} \text{ Bq}^{-1}),$$

$$T_s \quad = \quad \text{sampling time in min,}$$

$$Q \quad = \quad \text{sampling flow rate in m}^3 \text{ min}^{-1}, \text{ and}$$

$$T_f \quad = \quad \text{time factor given in Table 2.6.}$$

The relative standard deviation of the measurement in % can be calculated from the following equation.

$$\sigma_p = \frac{\sqrt{\dfrac{\text{gross count (cpm)}}{\text{time counted (min)}} + \dfrac{\text{background (cpm)}}{\text{time counted (min)}}}}{\text{gross counts (cpm)} - \text{background (cpm)}} \times 100 \qquad (2)$$

Example: An air sample was collected for $T_s = 5$ min, at a flow rate of $Q = 10$ L min^{-1} (0.01 m^3 min^{-1}). The counting was done from 35-45 min after the end of sampling. The total alpha count in that interval was 4200 counts. The counter background was 0.1 counts min^{-1} measured for a period of 25 min. The counter efficiency, E, was 0.48. T_f at 40 min, which is the midpoint of 35-45 min, is 7.21 obtained from Table 2.6. The total count in that interval was 4200 counts.

From Equation (1)

$$PAEC = \frac{(4200/10 - 0.1)}{(0.48 \ x \ 5 \ x \ 0.01 \ x \ 7.21)} = \frac{420}{0.173} = 2420 \ nJ \ m^{-3}.$$

The relative standard deviation of the PAEC measurement is

$$\sigma_p = \frac{\sqrt{\dfrac{420}{10} + \dfrac{0.1}{25}}}{420 - 0.1} = 0.015 \ (1.5\%).$$

2.2.4.5
MODIFIED TSIVOGLOU METHOD

The modified Tsivoglou method is more complicated than the Kusnetz, but it yields additional information: the concentration of individual radon progeny (Thomas, 1972).

A. Procedure.

The sampling procedure is identical to that used for the modified Kusnetz method. (Actually all three methods can be used with the same filter containing sampled radon progeny.)

After the end of sampling, remove the filter containing radon progeny from the filter holder and transfer it to the counting system for analysis. The standard counting intervals are 2-5, 6-20, and 21-30 min after the end of sampling. These intervals allow adequate time to transfer the filter into the counting system and record the counts from the different counting intervals. The technician or the person who performs the test is required to stand by for 30 min unless he uses automated counting equipment operated by a computer.

B. Calculation.

The equations needed to calculate the air concentrations from a 5 min sample and for the standard counting intervals are:

$$C_1 = \frac{[6.247\,N_t(2-5) - 3.028\,N_t(6-20) + 2.857\,N_t(21-30)]}{EQ} \qquad (3)$$

$$C_2 = \frac{[0.056\,N_t(2-5) - 0.776\,N_t(6-20) + 1.836\,N_t(21-30)]}{EQ} \qquad (4)$$

$$C_3 = \frac{[-0.832\,N_t(2-5) + 1.224\,N_t(6-20) - 1.389\,N_t(21-30)]}{EQ} \qquad (5)$$

$$C_p = \frac{[2.011\,N_t(25)\ 1.372\,N_t(620) + 3.954\,N_t(2130)]}{EQ} \qquad (6)$$

where

C_1, C_2, C_3 = the concentrations of ^{218}Po, ^{214}Pb, and ^{214}Bi, respectively (Bq m^{-3}),

C_p = the PAEC (nJ m^{-3})

$N_t(...)$ = the net counts in the intervals 2-5, 6-20, and 21-30,

E = fractional counter efficiency [(counts min^{-1}) /(disintegrations min^{-1})], and

Q = sampling air flow rate, L min^{-1}.

These coefficients were derived using 3.05 min for the half life of ^{218}Po (Martz et al., 1988). The half lives used for ^{214}Pb and ^{214}Bi were 26.8 and 19.9 min, respectively (ICRP, 1983).

The standard deviation equations which calculate the precision for each radionuclide are:

$$S_1 = \frac{1}{EQ}[39.0\,N_t(2-5) + 9.17\,N_t(6-20) + 8.16\,N_t(21-30)]^{1/2} \qquad (7)$$

$$S_2 = \frac{1}{EQ}[0.003\,N_t(2-5) + 0.062\,N_t(6-20) + 3.37\,N_t(21-30)]^{1/2} \qquad (8)$$

$$S_3 = \frac{1}{EQ}[0.69\,N_t(2-5) + 1.50\,N_t(6-20) + 1.93\,N_t(21-30)]^{1/2} \qquad (9)$$

$$S_p = \frac{1}{EQ}[4.04\,N_t(2-5) + 1.88\,N_t(6-20) + 15.6\,N_t(21-30)]^{1/2} \qquad (10)$$

where

S_1, S_2, S_3 =   standard deviation of the ^{218}Po, ^{214}Pb, and ^{214}Bi (Bq m^{-3}),

S_p =   standard deviation of the PAEC (nJ m^{-3}),

$N_t(...)$, E and Q were defined earlier.

The coefficients in the above four equations are the squares of the corresponding coefficients in Equations 3-6. Terms involving the uncertainty of the background count rate have been omitted because they are seldom significant.

A computer program based on the optimizing technique of Nazaroff (1984) (WWN5, for a PC; see Table 2.7) has been written to do the calculations in Equations 3-10. Results can be acquired within 15 sec after the end of counting. The program can also do these calculations from any three-count radon progeny measurement.

The two outlined counting procedures can be applied to the same filter sample simply by using two counting regimes in sequence: three counting intervals [(2-5), (6-20), and (21-30)] for the modified Tsivoglou method; a single counting interval from 35-45 min after the end of sampling (modified Kusnetz method). This provides a useful check against errors in transcribing data and in calculations.

2.2.4.6
RAABE-WRENN LEAST-SQUARES METHOD

As used here, the term "Raabe-Wrenn least squares method" refers to a certain configuration of alpha counters, a computer, hardware, and software for measuring radon decay product concentrations (Raabe and Wrenn, 1969). This sytem can handle up to 10 samples simultaneously, which is needed when measuring the particle size distribution of radon progeny particles (Sections 2.2.2.5 - 2.2.2.7). The method is also convenient for single samples.

A. Components.

1. Up to 10, as needed, drawer-type gross alpha counters, Model TH-29-B or equivalent.

2. A portable personal computer with two free 8-bit expansion slots.

3. Two CIO-CTR05 computer interface boards (Computer Boards 44 Wood Ave., Mansfield, MA 02048, 508-261-1123; FAX 508-261-1094) or equivalent, installed in adjacent slots of the above computer (item 2). One board must be set to address 0300 (hex) and the other to address 0304 (hex).

4. Two 37-pin D-connectors and hoods (Wire Pro, Inc., types 17-10370 and 17-1371; available from Newark Electronics, 212-963-0289). Install five female K-LOC connectors (King Electronics, Tuckahoe, NY, 914-793-5000) through the side panel of each hood, and wire them to the pins corresponding to the five CIO-CTR05 counter inputs. **Note**: the D-connectors must be made up in complimentary pairs so both can be connected to the CIO-CTR05 boards at the same time.

5. Up to 10, as needed, RG-174 coaxial cables with K-LOC connectors (King Electronics, Tuckahoe, NY, 914-793-5000).

6. Copies of the computer programs ALPHALOG (or ALPHALG2) and RWRENN6; see Table 2.7.

B. Procedure.

1. Set up apparatus; it is best to arrange the alpha counters in a line, five on either side of the computer. Install cables and power-packs.

2. Start the computer and make a directory with a name appropriate for the project you are working on. Copy **ALPHALOG** and **RWRENN6** into this directory.[1]

3. Start **ALPHALOG**; ensure that the computer's time and date are set correctly; when prompted, rn\un the test using the internal 1000 Hz pulser (a 10-second counting period is appropriate) to verify that the boards have been properly installed.

4. Skip over the "Enter parameters" step (it is usually more convenient to enter the parameters during counting).

5. Set the counting period (normally 60 seconds) and the number of intervals (normally 1000) and press **Y** in answer to the **"Ready to Start"** prompt. The program will go into a preview mode, in which counts are collected and displayed — but not saved — every 10 sec.

6. Collect the filter or particle size samples as described in Sections 2.2.2.5, 2.2.2.6, 2.2.2.7, or 2.2.4.2.

7. Promptly transfer the filters or other sampling substrates into the alpha counters; be sure that the substrates are placed "dirty side up" and ZnS scintillator disks are placed "shiny side up."

8. Verify from the computer preview screen that all channels are collecting counts; then with an eye on the stopwatch, start official counting by hitting **ENTER**. Make note of the time at which counting was started.

9. Once the counting is well underway, choose option **[F7]** to record the parameters associated with the sample. Enter information when prompted.

---

[1] *Or copy these two programs into a directory entitled* EOKPROGS *and place* EOKPROGS *into the* PATH *statement.*

10. After sufficient data has been collected, press **[Shift-F10]** to stop; 40 min of data is sufficient for calculating radon progeny, but 240-300 min is needed for calculating thoron progeny.

11. If the parameters were entered during the data collection period (step 9), press **Y** to proceed directly to the program **RWRENN6**, which calculates the decay product concentrations. If not, press **N**, then correct the parameters using an edit program such as MSDOS EDIT. Then run **RWRENN6**.

REFERENCES

George, A. C.
"Measurement of Airborne Radon-222 Daughters by Filter Collection and Alpha
 Radioactivity Detection"
B. Seifert, H. J. van de Wiel, B. Dodet, I. K. O'Neill (editors)
in: Environmental Carcinogens - Methods of Analysis and Exposure Measurement -
 Indoor Air, WHO, IARC Scientific Publications No. 109, Lyon, Vol. 12, pp. 173-179,
 March (1993)

ICRP
Radionuclide Transformations: Energy and Intensity of Emissions.
Annals of the International Commission Radiological Protection, ICRP Publication 38,
Vol. 11-13 (1983)

Kusnetz, H. L.
"Radon Daughters in Mine Atmospheres. A Field Method for Determining
Concentrations"
Am. Ind. Hyg. Assoc. Q., 17, 85-88 (1956)

Martz, D. E., R. T. Harris, and G. H. Langner
"The Half-Life of ^{218}Po"
Report UNC-J-002-88, UNC Geotech, Grand Junction, CO (1988)

Nazaroff, W. W.

"Optimizing the Total-Alpha Three-Count Technique for Measuring Concentrations of Radon Progeny in Residences"

Health Phys., 46, 395-405 (1984)

Raabe, O. G. and M. Wrenn

"Analysis of the Activity of Radon Daughter Samples by Weighted Least Squares"

Health Phys., 17, 593-605 (1969)

Thomas, J. W.

"Measurement of Radon Daughters in Air"

Health Phys., 23, 783-789 (1972)

TABLE 2.4

DRAWER-TYPE ALPHA PARTICLE COUNTERS USED AT EML

Designation	TH-29-B	Greybox
Built by	EML	EML
Date of design	1981	1975
Photomultiplier tube diameter (cm)	5.0	12.7
Largest filter accommodated (cm)	5	10
Window material	None	None
Filter-to-window spacing (mm)	2	3
Typical efficiency (%)	48	48
Estimated dead-time each pulse (µs)	50	50

TABLE 2.5

ALPHA-EMITTING STANDARD USED AT EML

Identifying marks	#16
When obtained	Fall 1981
Where obtained	Oak Ridge National Laboratory
Active material	^{241}Am
Size of active deposit (mm)	18
Certifying agency	NIST
Certified activity (Bq)	-
Certified emission rate (alphas sec^{-1})	92.08
Mounting material	Stainless steel
Diameter of mount (mm)	25.4
Estimated ratio of emission to activity	0.504
Condition as of February 1988	Good
Efficiency multiplier	0.992

TABLE 2.6

TIME FACTORS FOR USE IN THE MODIFIED
KUSNETZ METHOD

Time after sampling[*] (min)	Time factor
40	7.21
42	7.02
44	6.83
46	6.63
48	6.44
50	6.25
52	6.06
54	5.87
56	5.67
58	5.48
60	5.29
62	5.10
64	4.90
66	4.71
68	4.52
70	4.32
72	4.18
74	4.04
76	3.94
78	3.75
80	3.61
82	3.51
84	3.32
86	3.17
88	3.03
90	2.88

[*]Midpoint of counting interval.

TABLE 2.7

COMPUTER PROGRAMS USED AT EML IN CONNECTION WITH
RADON PROGENY MEASUREMENTS

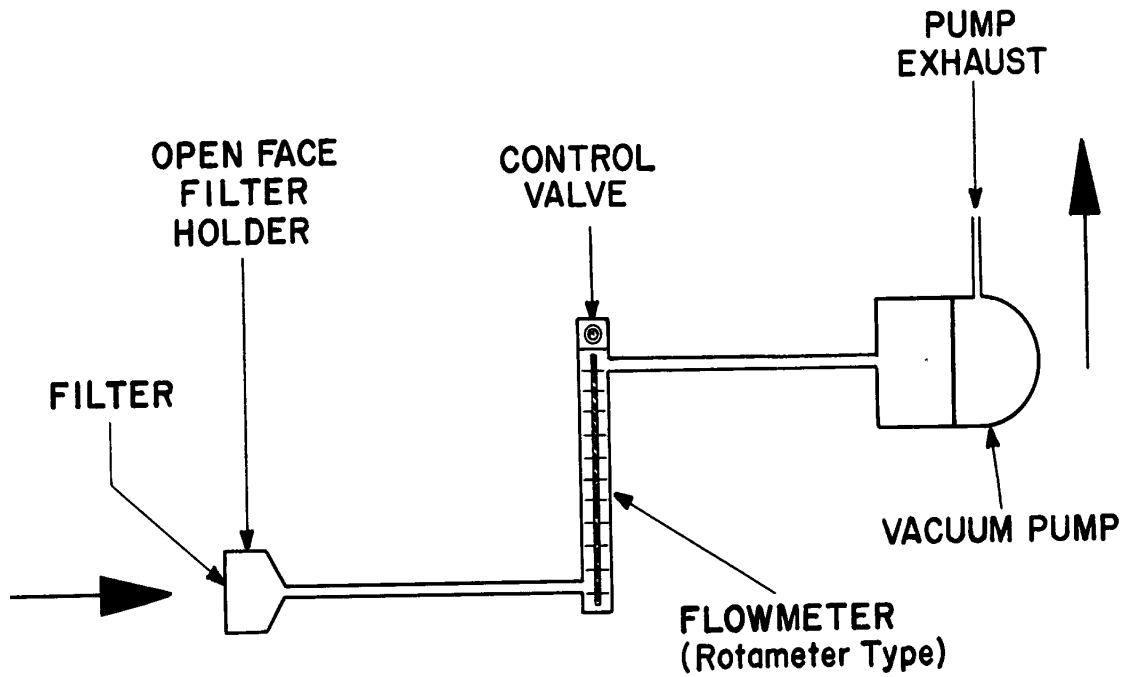| Name	Language	Machine	Method	Comments
ALPHALOG.PAS	Pascal	PC		Simultaneous collection of data from up to 10 alpha counters
WWN5.PAS	Pascal		Modified Tsivoglou	Nazaroff equations (1984) for any combination of sampling and counting times, including counting while sampling
RWRENN6.BAS	GW BASIC	PC	Least squares	Raabe and Wrenn (1969)

Figure 2.4. Sampling train for radon progeny measurements.

2.2.5 **ATMOSPHERIC TRACING**

2.2.5.1
INTRODUCTION

This section describes the apparatus and procedures used by EML to sample tracer gases used in studies of long range atmospheric transport. The analysis of the collected samples is described separately in Section 4.2. Atmospheric tracing is done primarily to test meteorological models that predict the atmospheric transport and dispersion of pollutants over distances as far as 3000 km from the source(s). Among other things, these models are used in assessing the potential for "acid precipitation" in different regions and under different meteorological conditions.

The tracer gases used in EML atmospheric tracing experiments are:

Tracer Gas	Acronym	Global Background $(fL\ L^{-1})$
perfluoromethylcyclohexane	PMCH	3.3
total perfluorodimethylcyclohexane	PDCH	28.8
perfluoromethylcyclopentane	PMCP	3.2
ortho(cis)perfluoro- dimethylcyclohexane	OC-PDCH	0.15
perfluorotrimethylcyclohexane	PTCH	<0.5

The first two tracer gases were identified by Lovelock (1982) and the others by Dietz (1987). These gases are nontoxic and chemically stable. They were selected because they have no natural or anthropogenic source apart from their release during tracing experiments, and because they are detectable at their background levels (shown above) using the procedures described here and in Section 4.2.

2.2.5.2
THE PROGRAMMABLE ATMOSPHERIC TRACER SAMPLER

The programmable atmospheric tracer sampler (PATS) (Model RD113, Gilian Instrument Company, Dawes Highway, Wayne, NJ 07470) is used as the primary sampler for all perfluorocarbon tracers. The unit, shown in Figure 2.5, is housed in a waterproof 36 cm x 25 cm x 20 cm container and weighs ~ 7 kg. It consists of two sections: the lid (air flow module, AFM) and the base (power control module, PCM).

The lid holds 23 sampling tubes, each containing 150 mg of Ambersorb adsorbent (Rohm and Haas). The Ambersorb absorbs the tracer from the sample air flowing through the tube. The air flow may be directed through a preselected tube by means of a multiple port switching valve (Scanivalve, 10222 San Diego Mission Road, San Diego, CA 92120) which is controlled by the PCM.

The base of the PATS contains a constant flow pump which draws air through the selected sampling tube. The constant flow pump is based on the EML tethered air pump system (Latner, 1986), and may be set to draw 10, 20, 30, 40 or 50 $cm^3 min^{-1}$. The base also contains programmable timer controls that allow for single or multiple start and stop times over a 7-day period. These controls are also used for automatic analysis when the unit is coupled to a gas chromatograph.

Two liquid crystal displays in the base indicate the time of day, the day of the week, and the tube number, and a digital printer records this information at each tube change. As a precaution against printer failure, this information is also stored in an integrated memory circuit module (Polito, 1987) mounted on each lid. The memory module is used during the gas chromatographic analysis phase (see Section 4.2), and this information is stored in a computer for inclusion into a data base.

Power is supplied by an internal rechargeable battery which can operate the unit up to 30 days. Longer periods of operation are possible if 110 V A.C. power is available. All lids are interchangeable and usable with any base, so that after 23 samples have been collected, a new lid may be substituted to continue sampling.

2.2.5.3
ADSORBENT TUBE BAKE-OUT PROCEDURE

A. Introduction.

Programmable atmospheric tracer sampler (PATS) adsorbent tubes are decontaminated (baked out) by resistance heating of each tube at $> 400ºC$. This method is applicable to all PATS lids. Up to 10 lids may be baked out at one time (Figure 2.6).

B. Special apparatus.

1. Constant current power supply, Power Mate Corporation Model BPA 2086-V or equivalent with special interface.

2. Micromaster programmable controller, Model No WP6201-AA-AA or equivalent.

3. Matheson Model 3800 gas pressure regulator or equivalent (two regulators required).

4. PATS bases and lids (up to 10).

5. Primeline (Soltec) two channel strip chart recorder or equivalent.

C. Special gases.

1. Nitrogen, ultra high purity (UHP) (99.999%) or equivalent.

2. Nitrogen, Matheson Purity (99.995%) or equivalent.

D. Procedure.

1. Attach each lid to each base whose batteries have been fully charged. Disconnect all battery chargers.

2. Insert a jumper in the receptacle labelled **Analysis Connector** of each base to disable the sampling pump.

3. Attach the UHP nitrogen line to the **To Equalization** port of each lid. Set the equalization nitrogen gas pressure to 207 kPa (30 psig).

4. Plug each lid AFM electrical connector into each base receptacle labelled **AFM Connector.**

5. Set the clock time and day of the week of each PATS base and power up each unit. The clock time is set at the thumbwheel switch marked **Set/Time Alarm** and the day of the week by placing the appropriate **Day Select** switch to **On.**

6. Manually cycle through each of the 23 tubes using the base manual **Valve Step** switch. Cycle each lid at least twice to assure seating of the Scanivalve. Leave the tube number of each unit at "00" and turn off the power to each unit.

7. Attach the Matheson Purity Nitrogen gas line to the **To PCM** connector of each lid.

8. Set the UHP nitrogen pressure to 138 kPa (20 psig) and turn the nitrogen gas on. Adjust the flow to ~ 30 cm^3 min^{-1}.

9. For each base, set the thumbwheel switch marked **Duration** to 0016 and the **Sample Quantity** switch to "24". This sets the switching time between tubes and the number of tubes to be sequentially decontaminated. There are only 23 tubes in each lid, however, the base must switch to the 24th tube in order to complete the bake-out cycle.

10. Plug all the electrical jacks into the **Desorption Power** lid connector.

11. Turn the constant current power supply on and set the current to 16.0 A.

12. Turn on the dual channel strip chart recorder to record the desorption voltage and current.

13. Quickly turn each unit on, push the PATS base **Operate** switch to **Off**, bring the **Alarm** toggle switch from **Manual** to **Multi** and then back to **Manual.** Bring the **Operate** toggle switch to **Run**. This procedure should be completed within 1 min for all 10 bases so that all units will start at the same time.

14. At the next minute, tube number 1 of each unit will switch into position, the current will be turned on and the tube will be heated to $> 400°C$ for a 15-min period. The contaminants will be desorbed into the nitrogen carrier gas stream. At the end of the 15-min heating period the current will turn off. At 16 min, the next tube will advance into position and the heating cycle will be repeated until all 23 tubes have been decontaminated.

15. Disconnect all wires and gas lines. Insert a 1/2 hole septum over all lid inlet and outlet ports and disconnect the lid plug from the base.

2.2.5.4
DEPLOYMENT, SAMPLING, AND RETRIEVAL

Information on the deployment, sampling and retrieval may be found in the report by Draxler and Heffter (1986), which gives a detailed description of one tracing experiment.

REFERENCES

Dietz, R. N.
"Overview of Perfluorocarbon Tracer (PFT) Technology Applied to Quality Assurance of ANATEX Tracer Data"
Anatex Model Evaluation Workshop, Orlando, Florida, Oct. 20 and 21 (1987)

Draxler, R. and J. L. Heffter
"Across North America Tracer Experiment (ANATEX), Comprehensive Plan"
National Oceanic and Atmospheric Administration, Air Resources Laboratory, Silver Spring, MD (November, 1986)

Latner, N.
"TAPS: Tethered Air Pump System"
USDOE Report EML-456 (1986)

Lovelock, J. E. and G. J. Ferber
"Exotic Tracers For Atmospheric Studies"
Atmospheric Environment, <u>16</u>, 1467-1471 (1982)

Polito, M.
"A Data Acquisition System for Atmospheric Tracer Studies"
USDOE Report EML-500 (1987)

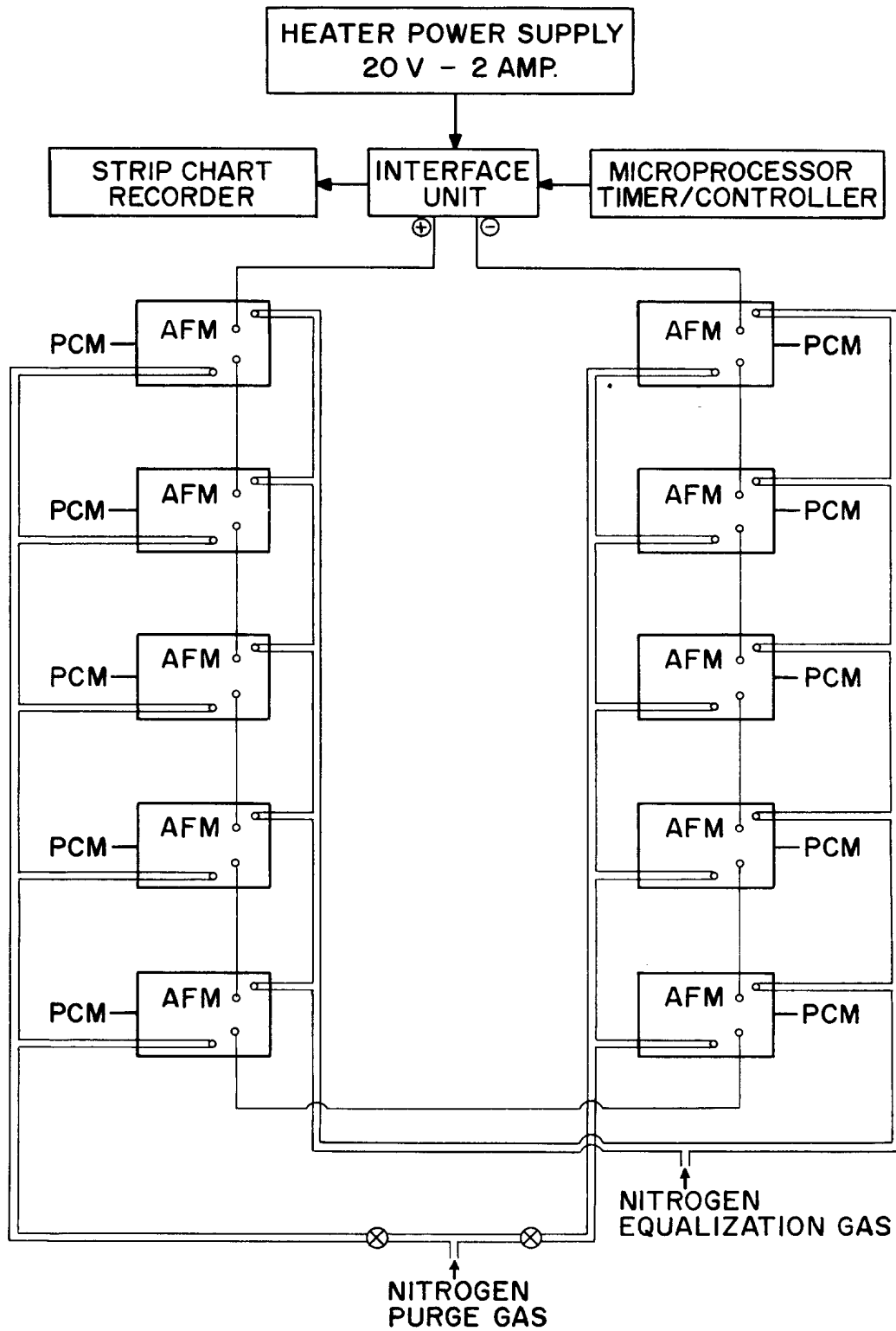Figure 2.5. The programmable atmospheric tracer sampler.

Figure 2.6. Configuration of the PATS lid absorbent tube bake-out system.

2.3 ATMOSPHERIC DEPOSITION

Contact Person(s) : Mathew A. Monetti

2.3.1 SCOPE

EML collects radioactive debris that is dispersed into the atmosphere from nuclear accidents and from the testing of nuclear weapons using two deposition collectors. These collectors enable us to measure radioactive deposition at locations far removed from the source. The collectors and collection methods described here have been used successfully for many years. Presently there are about 75 sites worldwide at which monthly collections are made. These samples are routinely radiochemically analyzed for ^{90}Sr. Following a known or suspected release of radioactivity, sampling and analysis protocol can be altered in order to determine the arrival time and composition of the debris.

We provide all the necessary supplies and instructions, in the appropriate language, and maintain written communication with site operators.

2.3.2 POT COLLECTOR

2.3.2.1
INTRODUCTION

The determination of ^{90}Sr and other radioisotopes contained in fallout can be carried out on the total material collected in a high-walled pot. The collection of fallout is simple, but the transfer of all the material in the pot must be done with great care in order to collect all of the radioactive material and to avoid a buildup of contamination in the pot, which may influence a subsequent sample.

2.3.2.2

APPARATUS

EML's collectors are stainless-steel pots, 0.076 m^2 in area, but many other high-walled vessels are equally suitable. The requirements include smooth surfaces that do not absorb the radionuclides and rounded corners for easy cleaning and durability. The pot collector is shown in Figure 2.7.

2.3.2.3

DEPLOYMENT, SAMPLING, AND RETRIEVAL

For our program, the pot collector is placed in an open area away from buildings or overhanging trees and shrubbery. An acceptable location is atop a one or two story building with a flat roof not shaded by other buildings or trees. The pot is placed outside on the first of the month and left there for one month. (During months of heavy rainfall, it may be necessary to remove and evaporate some of the water prior to the end of the month.)

At the end of the month, the water in the pot is evaporated to ~ 200 mL and transferred to a 1-L polyethylene bottle. Distilled water is added to the pot and, using a rubber spatula, the sides and bottom of the pot are scrubbed. The slurry is transferred to the bottle and the washing is repeated with 1:1 HNO_3, again scrubbing and transferring the slurry. Then the pot is washed a final time with distilled water and scrubbed. It is extremely important to transfer all the solid material in the pot to the bottle. If necessary more than one polyethylene bottle may be used for each month's collection. The bottle(s) should be carefully labeled with the site location, sample dates, and the monthly precipitation amount from the nearest recording station.

2.3.3 **ION EXCHANGE FALLOUT COLLECTOR**

2.3.3.1
INTRODUCTION

Like the pot collector, the ion exchange collector is designed for measuring fallout at sites removed from the source. At sites where personnel and facilities are at a minimum, the ion exchange collector is used. The collectors are exposed for monthly intervals and the collected fallout may then be shipped to a laboratory for analysis. The collector can be operated without a laboratory facility and consists of a funnel, an ion-exchange column, and a leveling device (all constructed of polyethylene) mounted in a wooden housing. The ion-exchange column is packed with paper pulp, ion-exchange resin and a glass wool plug, and is saturated with water. A utility light fixture with a 100-W bulb and a 30-m heavy duty extension cord is provided to heat the device in cold climates. The ion exchange fallout collector is shown in Figure 2.7. A descriptive diagram of the collector is shown in Figure 2.8.

2.3.3.2
MATERIALS

1. One plywood housing
2. One 30-cm polyethylene funnel
3. Two loaded resin columns, with caps
4. Two tapered caps
5. One leveling tube
6. One threaded T-connection
7. One piece of plastic tubing
8. One wash bottle
9. One set spare caps for mailing
10. One 100-W heating unit, where necessary

2.3.3.3
PREPARATION

The polyethylene ion-exchange column contains glass wool, paper pulp, and cation exchange resin. The glass wool is the standard laboratory material. The paper pulp is prepared from Whatman No. 41 filter paper or S&S No. 289 paper pulp by mixing in a blender with distilled water.

Dowex 50W-X12 cation exchange resin (Bio-Rad Laboratories, Inc., 1000-T Alfred Nobel Dr., Hercules, CA 94547) 50-100 mesh in hydrogen form is used. The resin is washed with deionized water.

Glass wool is added to the bottom of each column and cation exchange resin and paper pulp are added in that order. Approximately 17 cm of glass wool, 50 mL of wet settled resin and 4 cm of paper pulp are packed into a column filled with distilled water. Care is taken to prevent the formation of air pockets.

2.3.3.4
COLLECTOR LOCATION

The collector should be located with the same criteria used for selecting a rain gauge site. These criteria are generally that the collector should be able to receive precipitation in an open area free of buildings, trees, and other obstructions that might shelter the collector. In many locations a flat roof is available - this is a suitable location, when the collector cannot be placed in an open area at ground level.

2.3.3.5
SITE OPERATOR INSTRUCTIONS

1. Remove the ion-exchange column from its mailing tube and retain the tube for return of sample.

2. With the ion-exchange column inverted (small end up), remove the small cap and replace with the open tapered cap.

3. Force the short end of the J-shaped tube over the tapered cap and tape the other end to the column so that the small flexible hose is approximately level with the top of the tube.

4. Turn the column upright (large end up) and remove the large cap. Screw the column to the polyethylene funnel and place in the wooden housing. (Retain both caps for return of sample.)

5. Lead the long flexible tube, which serves as an overflow, through the hole in the side of the housing.

6. If subfreezing weather is anticipated during the month, hang the heating element from the eyelet in the housing and switch on during periods when snow or ice may accumulate in the collector.

7. The collector is left exposed continuously for 1 month, beginning on the first day of the month.

8. It is important that the resin column be kept saturated with water. If a dry period extends for several days, examine the column and add water if necessary. A plastic wash bottle is provided for this purpose. In most areas, tap water is satisfactory. However, in a few localities where rain water is collected directly for household use, distilled water should be used.

9. At the end of the observation period, use the wash bottle to rinse down any dust that has accumulated in the funnel and into the column, and wipe the inside of the funnel thoroughly with a wet tissue, which is then added to the column.

10. Remove the ion-exchange column from the assembly and replace the caps. (The new column can be installed at this time.)

11. **Note**: The column label should contain the station name, the month of exposure, and the total amount of precipitation recorded by the station during the month.

12. Pack the column in the original mailing tube and return to the analytical laboratory.

2.3.4 WET/DRY DEPOSITION COLLECTOR

2.3.4.1
INTRODUCTION

We examine the wet phase and dry phase of deposition to further understand atmospheric pollution deposition processes, and the physical and chemical characteristics of these pollutants. Our measurements of fallout from weapons tests were largely directed towards stratospheric fallout, which is primarily deposited by way of precipitation scavenging. Dry deposition processes are also important and, therefore, it is desirable to measure wet and dry deposition separately.

2.3.4.2
APPARATUS

EML's wet/dry deposition collector is a two-bucket system with a movable peaked cover designed to expose the wet and cover the dry bucket at the onset of precipitation, and vice versa (Volchok and Graveson, 1976). There is a conductivity sensor on each side of the roof. The sensor base plate electrode is separated from the parallel bar electrode above by a thin (about 0.8 mm) air space. Any electrical flow between the electrodes activates the motor in the base of the unit and the counter-weighted lid moves to cover the dry bucket. When the circuit is broken, the cover returns. Heaters are mounted below the sensors and are activated when the temperature drops below 4℃ to melt snow or ice accumulations on the sensor, or when the instrument is in the wet collection mode to evaporate moisture from the sensor. Thus, when properly adjusted, the wet collector will be exposed only during periods of continuous precipitation.

2.3.4.3
SAMPLING AND DEPLOYMENT

In the past, we have used stainless-steel pots to collect radioactive debris from weapons tests, and heavy polyethylene buckets to collect trace metals. Since many of EML's sites are remote from the Laboratory, we have adopted a commercial container which has a positive closure to prevent leakage during shipment. The opening in the plastic bucket is about 30 cm in diameter. The sampling containers for these wet/dry collectors are polyethylene buckets particularly well-suited for shipping because of the design of the lip and cover [Freund Can Co., 167 West 84th St., Chicago, IL 60620 (As-1137/5541 plastic pail with cover)]. On the underside rim of the cover there is a groove with an O-ring seal. When the cover is securely hammered onto the bucket with a rubber mallet, no leakage will occur. The cover is destroyed when it is removed from the bucket at the Laboratory. The sample buckets are shipped in fiber mailing boxes.

In 1978, EML started to use Aerochem Metrics Model 201 wet/dry collectors (Bushnell, FL) at some of our sampling sites. This collector is similar in concept and design to EML's collectors. Both the EML and the Aerochem Metrics collectors are shown in Figure 2.9. The major mechanical difference is the Aerochem Metrics clutch system and switching system. EML's collector uses a shear pin to prevent damage to the motor when the lid is frozen in place. The Aerochem unit has a clutch on the motor drive of the covering lid. This clutch eliminates the problems caused by frequent shear pin breakage common to the EML units at windy or cold sites. The switches on the motor box of the Aerochem Metric unit are mercury-wetted switches, whereas microswitches are used in our units.

REFERENCE

Volchok, H. L. and R. T. Graveson
"Wet/Dry Fallout Collection"
in: Proceedings of the Second Federal Conference on the Great Lakes, Great Lakes
  Commission, pp. 259-264 (1976)

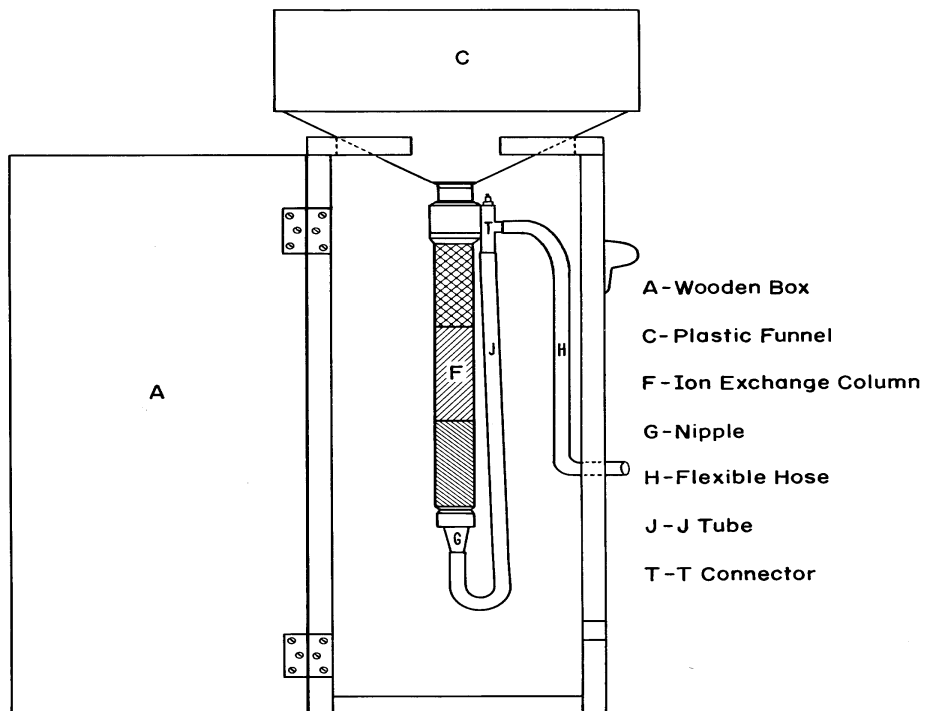Figure 2.7. Ion-exchange fallout collection and fallout pot collector.

A - Wooden Box

C - Plastic Funnel

F - Ion Exchange Column

G - Nipple

H - Flexible Hose

J - J Tube

T - T Connector

Figure 2.8. Ion-exchange fallout collector.

Figure 2.9. Aerochem metrics wet/dry collector and EML wet/dry collector.

2.4 SOIL

Contact Person(s) : Catherine S. Klusek

2.4.1 SCOPE

Presented in this section are possible uses of soil sampling and some recommended procedures for this sampling and for the preparation of the samples. An attempt is made to point out some of the problems that exist in site selection, and the sampling methods that are available.

2.4.2 SAMPLING OBJECTIVES

2.4.2.1
INTRODUCTION

The sampling of soil is a useful approach to determine the accumulated amounts of airborne long-lived radioactive and stable contaminants that deposit on the ground. Soil sampling is, however, of questionable value in attempting to estimate small increments of deposition over a period of a few years or less. It is not recommended as a routine method of environmental monitoring except in preoperational surveys.

Historically, soil sampling procedures for radionuclides were modifications of techniques used in agriculture and engineering. Often, not enough emphasis is placed on the importance of a proper sampling method to accurately represent the total pollutant being sampled. The objectives of the project should be well-defined and the degree of precision required should be established before sampling. The purpose of the project (deposit, resuspension, root uptake) should dictate the type of sampling (total inventory, surface sampling, depth profile) used. Site characteristics, such as soil type, topography, source, and current distribution of the contaminant must be taken into account when designing the study. Other factors to be considered in the design stage are the uniformity of the contamination, the required accuracy necessary to provide reasonable results, and the minimization of cross-contamination.

2.4.2.2
DEPOSITION INVENTORIES

The most direct use of soil measurements is for estimating the inventory of the material deposited over a given area. EML has used soil analyses in estimating the deposition of ^{90}Sr (Hardy et al., 1968) and plutonium (Hardy et al., 1973; Krey et al., 1976a) on a worldwide basis, and in estimating the deposition of ^{90}Sr (Hardy et al., 1972), ^{137}Cs (Beck and Krey, 1980) and plutonium (Hardy, 1976) northeast of the Nevada Test Site. Such inventories require the selection of a sufficient number of representative sites, with the density of the sites depending on the accuracy sought. A statistical sampling scheme is generally not employed in sampling global fallout because the aerial distribution and particle size are nearly uniform over large areas (i.e., latitude bands). Low-level baseline sampling would require the careful selection of sites that serve to integrate fallout over time.

The most useful measures of the concentration of deposited material in soil relate to the amount per unit area. Sampling is therefore carried out in such a way that the weight of the material collected can be directly related to the area sampled and the depth of the sampling. The analytical results from a weighed aliquot of the soil sample can then be readily related to area concentration.

An additional requirement is that the sample is taken to a sufficient depth so that all of the deposited material is sampled. Without previous knowledge of the depth of penetration, an excessive sampling depth must be selected. This results in dilution of the radionuclide or other contaminant of interest. If time and cost considerations allow, it is recommended that a depth profile be taken (see Section 2.4.3.3). An optimum depth for sampling, which should contain 90-95% of the total material of interest, can then be selected. In our worldwide ^{90}Sr network we have increased our depth of sampling over the years from a few centimeters to 30 cm. Even so, at some sites typified by calcareous soils, the ^{90}Sr has moved deeper than 30 cm. Sampling parameters should be reevaluated and decisions made on a site basis.

Criteria for site selection. When the accumulated deposition over a given time period is to be estimated by soil sampling, it is necessary that the area selected for sampling has been undisturbed for at least the time interval that is of interest. As the time

interval increases, it becomes increasingly more difficult to obtain an accurate history of the site. In studies of cumulative fallout deposition, areas undisturbed since the early 1950s are sought. Institutional property will have certain advantages over private property in these cases.

The second criterion, that of representativeness of the sample site, depends on the surroundings and the meteorological or climatological factors of the area. This generally requires that the site selected should be at the center of a large, flat open area. Accumulative areas at the foot of slopes, in low spots or in flooded areas are not suitable. The site should not be near enough to buildings or trees to be sheltered during blowing rains. The sampling location should be 100 m or more from dusty roads. A large area of collection is desirable to make the sample more representative. A surface area of 500-1000 cm^2 is adequate when a composite of 10 cores is taken over a reasonable distance (~ 30 cm apart).

The third criterion, that the deposited material remain in place, generally requires that the area be vegetated and have moderate to good permeability. There should be little or no runoff during heavy rains and no overwash at any time. The soil should have a base exchange capacity adequate to keep the contaminant from being readily leached into the ground water. A good grass turf aids in absorption of water and reduces the likelihood of runoff. Such sites are frequently found on smooth ridge crests, level virgin land, and in well-kept lawns and grounds around institutional buildings.

At EML, we recommend short-cropped grass sods as the most suitable sampling areas. Higher stands of vegetation may bias the collections by acting as a filter to remove airborne material in excess of what would normally be deposited. In wooded areas, the nonuniform distribution of tree canopies will lead to unevenness in deposition of fallout materials. Soils having high earthworm activity should be avoided because of uneven mixing of the soil to considerable depths. Rodent activity also makes an area unsuitable for sampling (Alexander et al., 1960).

2.4.2.3
DEPOSITION INCREMENT

Incremental estimation of a contaminant through soil sampling and analysis should be undertaken only after careful planning. Because of the large uncertainties in sampling and the inherent variability in soil, estimating short-time increments of deposition history or deposition changes of small degrees is not recommended. When monitoring global fallout, short-term changes in radiation concentrations are generally small compared to the variability in the local radionuclide distribution. Direct collections of deposition or of airborne material are much more specific and yield more information with respect to the time when contamination occurred.

2.4.2.4
OPERATIONAL OR ACCIDENTAL RELEASES

Following an acute release of a contaminant or an accident at a specific facility, surface soil sampling soon after the event can be used to define the contamination contours or distribution pattern. This would require sampling only the top 5 cm of soil, including the vegetation. Our experience indicates that attempts to sample a shallower depth results in less reproducible samples. In many areas, a site meeting the criteria for an optimal sample has a root mat extending several centimeters into the ground, and it is rarely possible to remove a core <5 cm in depth intact.

In the case of accidental or operational releases, consideration must be given to liquid effluent as well as airborne particulate deposition. The amount deposited may vary with direction and distance from the release point. Airborne particle dispersal is affected by particle size and meteorological patterns. Gaussian plume models which take into account meteorology, stack height, topography, and deposition velocity of the particles are used to map local dispersion patterns. The horizontal dispersion of liquid effluent releases is influenced by the composition and quantity of the liquid, the topography, the soil type, and the properties of the contaminant of interest.

Sampling techniques used in evaluating acute releases are more site dependent and methods used for fallout deposition may not be appropriate. Differences in the methods

are dictated by the nature of the distribution of the contaminant in the soil, the range of particle sizes, and the generally higher levels of releases. Soil sampling in locally contaminated areas, such as Rocky Flats, can be inventoried by EML methods where the contaminant was initially made airborne in micron size particles from the source (Hardy and Krey, 1971).

Criteria for site selection. Wind roses and atmospheric dispersion calculations provide useful guidance in selecting appropriate soil sampling locations. Figure 2.10 shows a suggested distribution of sampling sites covering the area surrounding a plant, with emphasis on the downwind direction. This approach to soil sampling was used by EML at Rocky Flats (Krey, 1976; Krey et al., 1976b). Samples are commonly collected from a regular grid or a radial pattern, at least close to the facility. When using such a systematic sampling design, care should be taken to ensure that the repetitive spacing of the sampling points does not introduce an error in the results. Consideration should also be given to the likelihood that a grid point can not be used for sampling. Some alternative random location or selection procedure should be established during the planning stages. Soil sampling within a 16 km radius should give an adequate preliminary picture of levels around the plant. It is also suggested that one or more samples be taken close to the center of the most heavily populated area in the vicinity of the plant. When sampling at facilities that release activity via tall stacks, sample locations at considerable distance from the potential release point may be indicated.

The same site selection techniques can be used for a preoperational survey around a plant. When choosing sampling locations at this early stage, it is desirable to select areas that can be resampled at a later time, should it become necessary. Samples should not be taken from the identical location since the sampling depletes the soil and alters the concentration of the pollutant under investigation. Rather, a sampling plot of several square meters should be established and samples removed from different areas of the plot.

2.4.2.5
AGRICULTURAL AVAILABILITY

When evaluating soil for uptake availability of a contaminant of interest, it is not necessary to measure the total deposit but only the amount in the root zone that would be available to the plant or crop of interest. In most cases this would be the depth of the

plowed layer. In addition to root zone concentration, the extent to which the nuclide or contaminant is chemically available for uptake must be considered. The availability of the particular contaminant to the plant is rarely 100%. The processes influencing the mobility and availability of radionuclides in soil are complex and have been discussed by several researchers (Schulz, 1965; Russell et al., 1971; Eisenbud, 1987). Routine procedures for soil sampling will not be appropriate for this type of study because of the distribution of the nuclide. Field studies of environmental levels of radionuclides in agricultural soils have confirmed the nonuniformity of the concentration distribution due to mixing of the soil during cultivation and depletion by plant uptake and harvesting (Hardy and Bennett, 1977).

2.4.2.6
RESUSPENSION AVAILABILITY

There is no standard method to sample for availability of a contaminant in the soil for resuspension. The direct measurement of the airborne contaminant is the only sound approach to the problem of evaluating exposure to resuspended material. The practical problems of the mechanics of sampling make it difficult to take a very shallow soil sample with reliability. In trying to sample 1 cm or less it is very difficult to take into account the variation in the ground contour and to reproduce a particular sample. EML has tested other techniques, such as pressing gummed film to the surface, but these also have many mechanical difficulties (Krey et al., 1977). Another approach is to measure a depth profile at the site and plot the contamination as a function of depth. This has been found in practice to be an exponential, and extrapolation of the curve to zero depth might give an index of suspendability. This approach is limited by the error introduced if the shape of the curve as it approaches the surface deviates markedly from an exponential function. Resuspension concepts and prediction of the degree to which resuspension may occur have been reviewed (Healy, 1980; Sehmel, 1980).

2.4.3 **RECOMMENDED PROCEDURES**

A few soil sampling methods will be discussed in this section. Variations on these techniques or alternate methods (ASTM, 1983a; Bernhardt, 1976; EPA, 1979, 1983; Gilbert, 1987) may be necessary to accommodate site specific characteristics or the

objectives of the project. The standard EML procedure for collecting soil is described in the section on core method below, i.e., 10-8.9 cm diameter cores spaced 0.5 m apart on a flat grassy lawn are collected to a depth of 5 cm, representing an area of 620 cm^2.

In all the procedures discussed, it is important to consider the potential for cross-contamination. If depth profiles are taken, it may be necessary to decontaminate or discard tools as different levels of contamination are sampled.

2.4.3.1
CORE METHOD

The procedure described here is designed to obtain samples that will measure the total amount of an initially airborne contaminant that has fallen out in a given area.
The criteria for selecting an optimal site have been discussed in Section 2.4.2.2.

All analytical values must be related to the surface area sampled. The surface area and depth define the volume; the weight of the volume of dry soil defines the field bulk density. These data are necessary to convert to radioactivity concentration per unit surface area (i.e., Bq m^{-2}).

A recommended procedure is described as follows.

1. Following the selection of an undisturbed site which meets the criteria previously discussed, lay out a straight line transect about 4.5 m long. If the site is to be re-sampled at a later time, record distances to fixed landmarks to identify the relative location of the transect or adopt a systematic scheme or grid.

2. If the vegetation cover is not to be included with the soil sample, or is to be kept as a separate sample, the vegetation is removed to the surface level.

3. Using the 5 cm depth top soil cutter, press it into the ground without twisting or disturbing the grass cover or surface soil. This may best be accomplished by stepping on the rim of the cutter with both shoe heels. If more force is required, a rubber mallet may be used. Gently twist the handle of the cutter to cleanly remove the top soil plug. Place the core in a plastic sampling bag.

4. Repeat the process until the desired number of cores have been sampled. We
    recommend 10 cores for providing a representative sample (Alexander et al., 1961).
    Samples of 20 and 10 core composites were collected in about 10 areas and no
    significant differences were found in the estimated ^{90}Sr deposit. Compositing soil
    samples provides a larger sample volume and possibly a more representative sample
    of the area. Compositing is discussed further in Bernhardt (1976) and Gilbert (1987).
    Take 10 top soil cores in a straight line about 30 cm apart, placing the cores in a
    plastic bag. (The total area sampled is 620 cm^2.)

5. Sometimes it may not be possible to remove a 5 cm depth plug cleanly because of a
    thick root mat. If the top soil and bottom soil are to be combined, a 10 cm or 15 cm
    deep cutter may be used to remove the top soil by pounding it part way into the
    ground with the rubber mallet, until it is possible to remove the core intact.

6. Next, take the subsoil samples down to the desired depth with the auger. A slight
    downward pressure with slow turning will guide the auger. When the cylinder is
    about 3/4 full, remove the auger slowly and either tap out or scrape out the soil with a
    large flat blade knife. Continue to use the auger until the desired depth has been
    sampled. If rocks or roots impede the auger, it may be possible to carefully remove
    them. They should be included with the sample. If, however, this destroys the core,
    the sample should not be used. It is a useful practice to place the soil from the core as
    it is removed into a plastic pail until the entire depth is removed. Then, if the core is
    not suitable, it may be poured back into the hole. Only after the entire sample is
    successfully removed is the soil added to the sampling bag. Repeat the procedure for
    the remaining cores.

7. After collection, label the plastic bag containing the sample, fold, and seal with a
    heavy duty stapler. If a portable scale is available, the wet weight can be taken in the
    field. Then place the sample in a canvas bag and tie firmly. The label should include
    the date, location, and depth.

8. The holes should be filled with top soil to prevent an accident. A new grass cover
    will develop in a few weeks. If immediate restoration is necessary, plugs may be cut
    from a piece of sod.

Using the above procedure, a site can be sampled in two increments, 0-5 cm and 5-30 cm. This is useful in areas where most of the contamination is in the surface cut of the soil. In other sampling situations using cores of 10 and 15 cm depth will provide incremental samples: 0-5, 5-10, 10-15, and 15-30 cm. When attempting this type of incremental sampling, attention must be given to two sources of error: contamination by fall-in of soil from the upper layers of more highly contaminated soil as the subsequent cores are taken from the hole and error in depth due to compaction.

A. Limitations.

Implicit when using the above sampling method is the need to take the sample deep enough so that all the radionuclide deposited is collected. The extent of vertical penetration will depend primarily upon the soil type with other factors, such as amount of annual precipitation and chemical form of the nuclide, playing a confounding role. [See Section 3.3 Field Gamma-Ray Spectrometry for a discussion of the use of in situ spectrometry to aid in establishing the depth of penetration.]

There is no simple satisfactory way of sampling powdery, dry, loose, single grain soils by this core method. It is best to take samples when the soil has enough moisture to be coherent even if this requires wetting the area to be sampled by sprinkling. An alternate method for sampling loose soils is to leave the corer in place and scoop out the contents. Only one composite depth can be taken however, since once the corer is removed the integrity of the core is lost.

B. Sampling equipment for the core method.

Tools for sampling may be of any material and type that will take a core of equal area through its entire length. The following equipment are used at EML:

1) Barrel auger - Standard Type No. R-HEO, 8.2 cm ID, with T-handle, Arts Machine Shop, American Falls, ID.

2) Top soil cutters - 5, 10, 15 cm depth, 8.9 cm ID. Made from 0.155 cm thick cold-rolled steel. One end sharpened on a lathe, the other end fitted with a welded handle.

3) Other equipment -

Long, flat blade knife for removing cores from auger

Measuring tape (~ 8 m)

File for sharpening top soil cutters

Rubber mallet

Plastic bucket (5 L).

2.4.3.2
TEMPLATE METHOD

Although the core method is preferred, there are areas where the rocks will make it impossible to use this technique. An alternate method we employ in these areas is to cut out a 900 cm^2 sample using a 31.6 cm square-template for guidance. The soil and rocks are removed with chisels and scoops down to the desired depth. The rocks are included and weighed with the sample. The large rocks can be discarded after removing loose dirt. The remaining smaller rocks are crushed as part of the sample.

This method is comparable to the ring method used by the Nevada Applied Ecology Group (NAEG) for sampling sandy and rocky soils. Here a 12.7 cm ID ring with a lip to assure constant depth of penetration is used. The soil adjacent to the exterior of the ring is removed to the depth of the ring. The confined volume of soil is then transferred to a plastic bag using an appropriate tool. Depth profile may be drawn using this method by repeating the steps for each subsequent depth to be sampled. Removal of soil exterior to the ring is necessary to minimize the possibility of cross-contamination of the deeper, less radioactive fractions (Fowler et al., 1974). A minimum of five separate samples (633 cm^2) are recommended along a transect and composited for analysis.

A. Sampling equipment for the template method.

Tools for sampling may be of any material that will maintain a rigid shape and straight edge. The sampling equipment used at EML consists of:

1) A square template 20 cm or 30 cm on the inner edge made of 0.157-cm thick cold-rolled steel, with holes at the corners,

2) Pins or long nails of corresponding diameter to the corner holes of the template to anchor the template to the surface,

3) Chisels, knives, and small shovels.

2.4.3.3
TRENCH METHOD

The trench method is used to determine the depth of penetration of a radionuclide or contaminant or to establish a detailed depth profile. The most suitable area for taking soil profiles is one where there are no rocks and stones, and very few pebbles. The procedure works well in sandy loam, loam or loamy sand types of soil (Hardy, 1974; Fowler et al., 1974).   Use of this method in unsuitable areas will result in cross-contamination of lower profile cuts with higher specific activity subsoil from upper layers. Cross contamination effects of worms and burrowing insects, cracks from swelling, and other biological and physical disturbances must be considered.

A recommended procedure is described as follows.

1. As far as grass cover and terrain are concerned, the site selection criteria previously described apply. As in the core sampling method, the depth profile samples are taken so that the weight and depth of the material collected can be directly related to the area.

2. If the vegetation represents a seasonal growth, it should be clipped to 2.5-5 cm over a measured area.

3. Lay a tarpaulin (about 0.6 m^2) on the ground near the clipped area. Dig a trench of an appropriate size for ease of access (about 60 cm wide by 90 cm long by 60 cm deep, about 15-25 cm deeper than the desired sampling depth) immediately adjacent to the clipped area, placing the dirt on the tarpaulin. Usually the sod can be cut out in blocks making it easy to replace after sampling.

4. The face of the trench (adjacent to the clipped area) is smoothed from side to side with a flat blade shovel or mortar trowel, making it perpendicular to the surface.

5. A metal flat-bottomed three-sided pan with sharpened edges on the open side (15 cm x 15 cm x 5 cm deep; 230 cm^2) is pressed into the face from ground surface to 5 cm. Remove the first cut and seal in a small plastic bag.

6. Cut away the top soil on either side of the cut to make a shelf about 35-cm long by 15-cm wide and 5 cm deep from the surface. Lightly brush away any particles that may have fallen on the shelf.

7. Again, push the open-end cutting pan into the side and cut out the next incremental sample. Continue this procedure until the desired depth is reached. The actual depth of each cut can be determined by placing a two by four on the surface and measuring to each subsurface.

8. When all the samples have been taken, fill the trench with dirt on the tarpaulinand replace the sod taken from the trench.

A. **Limitations.**

A depth profile is only useful for finding the relative vertical distribution of the radionuclide. Since only 100-300 cm^2 of surface area at one spot is sampled when taking depth increments, the integrated deposit is not necessarily representative of the area.

The trench method is more time consuming and more difficult than taking core samples. Therefore, researchers rarely sample and composite more than two samples per trench and rarely take duplicate profiles. However, if care is taken, there will be very little cross contamination and the data collected in terms of the depth profile will be more accurate.

Some consideration should be given to the problems introduced by sampling across soil horizons. Soil horizons differ in chemical and physical properties.

B. **Sampling equipment for the trench method.**

The sampling equipment used by EML consists of:

1) Three-sided square pan with cutting edges on the open side (15 cm x 15 cm x 5 cm deep made of 0.157 cm thick cold-rolled steel, welded at the corners);

2) Mortar trowel;

3) Long flat-blade knife;

4) 1.2 m piece of two by four.

2.4.3.4
NONHOMOGENEOUS TERRAIN METHOD

Previous site selection techniques discussed above assure a representative sample by the homogeneous and undisturbed nature of the location. In geographic areas characterized by sparse vegetation, wind erosion, and nonuniform drainage, the fallout is

expected to be nonuniformly distributed. A procedure for recovering a representative soil sample from complex terrain for the purpose of determining the integrated radionuclide inventory deposited in the area is described in Krey and Klusek (1995). The following protocol is used in such a nonhomogeneous terrain area.

1. In the area of interest, select a site that is visually representative of the area. An optimal area would be without excessive rills, washout areas, and gullies that would indicate areas of intense erosion. If the area is very diverse in landscape, consideration should be given to subdividing the area and taking samples in each area.

2. Determine the number and types of covers (strata). Some common strata are: open pavement, trees, shrubs, grass, rocks. Some judgment is required with small trees and large bushes. Vegetation should be assigned as a cover type if there is a significant difference in the character or amount of accumulation of soil at their base. It is preferable to preassign the vegetation to a category before attempting to estimate the linear percentage to avoid changing the category's characteristic in midcount.

3. Select, at random, a starting point and mark off a 100 m straight line transect in a randomly selected direction.

4. Using a measure that is a reasonable small unit, i.e., every 0.1 m, count off the number of meters intersected by each stratum. Estimate the proportion of each cover along the total transect.

5. Determine the number and method of sampling to be used in each identified stratum. Samples should be taken in every stratum that represents more than 5% of the total transect. For each cover type to be sampled:

   a) Sample (systematically) by the core method at a fixed interval (e.g., 6 m) along the transect using the closest sampling location within ± 0.5 m of the mark.

   b) Sample (randomly) each cover type by the template method. It is only feasible to take two to four samples by this method.

6. The samples taken in a stratum are combined. At this point the soil samples (one per stratum) can be analyzed:

   a) independently to obtain information pertinent to each stratum and the total inventory estimated by summing the proportional contribution from each stratum;

   b) the soil samples can be composited based on the areal proportion and only this composite sample for the site is analyzed.

2.4.4 **SAMPLE PROCESSING**

The procedures to be followed to process a soil sample to obtain a representative subsample for analysis depend to some extent on the nuclide of interest, the size of the sample, and the amount of processing already undertaken in the field. Any one of, or a combination of, the following procedures may be employed. The soil may be crushed to reduce the size of stones, sieved to remove sample content above a desired size, blended to obtain a more homogenous distribution of particle sizes, or milled to reduce the particle size of the soil. If the sample was sieved or split in the field or a small sample was taken, the preparation process may be eliminated. For some purposes, it is possible to remove large nonporous stones and not grind them to size, but they must be weighed separately and an appropriate allowance made. As a general rule, at EML samples are air-dried, crushed to break up large rocks, blended to allow a representative aliquot to be removed, and only this subsample is pulverized. The pulverizing reduces the soil to a standard particle size. Other preparation protocols are addressed in ASTM (1983b).

Global fallout is relatively homogeneous in particle size and distribution in the sample. When sampling accidental or operational releases, the procedures described here may not be adequate. Care must be taken that the subsample taken for analysis accurately represents the total sample. This will depend on the size and the degree of heterogeneity. Multiple subsampling and multiple analysis may be the only technique available to adequately define the content of radionuclides in heterogeneous samples.

Care must be taken in all stages of processing to avoid contamination from previously prepared higher concentration samples. In addition to careful cleaning of the equipment

between samples it is sometimes advisable to run blank samples (e.g., sand) between the samples being processed. Another technique to minimize cross-contamination is to order the processing of samples starting with the lowest level samples first. This is difficult to know in an exact sense, but generally deeper depth samples will have a lower content than surface samples.

<div align="center">

2.4.4.1
DRYING

</div>

A recommended procedure is described as follows.

1. The "excludable" fraction of the sample should be defined, dependent on the contaminant of interest. Vegetation, root mat, large organic pieces, stones, etc., in some cases can be discarded. If this is the case, the sample can be screened using a 6 mm sieve or a 10 mesh screen. The appropriate steps of drying process (e.g., Step 3 and/or 5) can then be skipped.

2. If facilities are available, the samples can be dried at 100℃ overnight. Oven drying has the advantage that most materials will become brittle enough to mill properly. Otherwise, spread out the sample on a plastic sheet or in trays and allow to air dry. This will take 3 days to 2 weeks. Turning the soil will facilitate the drying process especially when the drying area does not allow the soil to be spread in a thin layer. However, turning is not advised when the rising dust might cause contamination of other samples.

3. Break up soil aggregates and pull apart the top soil plugs consisting of vegetation and root mat. Using scissors or clippers, cut up the vegetation so that it is distributed homogeneously throughout the sample.

4. When completely dry, weigh the entire sample.

5. Remove large rocks, weigh separately, and discard.

2.4.4.2
HOMOGENIZING

When using equipment in this stage of preparation close attention must be paid to cross-contamination. Machinery should be dismantled and decontaminated between samples.

1. If the sample contains small rocks or pebbles:

   a) crush the entire soil sample, reducing the sample to 6.35 mm, or

   b) sieve the entire sample through a 12.7-mm screen.

2. Then blend the entire sample for 15-30 min.

3. Spread out the sample, mark off quarters, and take scoop-fulls from each quarter in a consecutive manner until about 3 kg has been collected.

4. Pass this subsample of soil through a grinder, ball mill, sieve or pulverizer. The pulverizer used at EML reduces the soil to <1.3 mm (15 mesh equivalent). Transfer to a wide-mouth polyethylene bottle.

5. When ready for analysis, roll the bottle to mix the sample thoroughly.

2.4.4.3
RAPID PREPARATION METHOD

An alternate soil sample preparation procedure is employed to allow rapid processing and aliquoting when gamma-ray spectrometric analysis for short-lived radionuclides is desired. This procedure was developed to provide timely data from surface soil samples collected following the Chernobyl reactor accident. After weighing the sample in the wet state (as received), the entire sample is sieved through a steel mesh screen with square openings, 1.27 cm on edge. Rocks and pebbles of greater size are brushed and discarded. Vegetation and root mat are cut to a size (about 0.6 cm) that permits them to pass through

the sieve. The mixture of wet soil and vegetation is homogenized by hand for several minutes before aliquots of about 100 g are removed. If desired, the remainder of the sample can then be dried and processed as previously described.

In experiments involving 75 sets of duplicate wet samples, 87% of the samples agreed with each other to within two standard deviations of the propagated Poisson counting uncertainties. This indicates a reasonable degree of homogenization of the wet sample. In addition, the ratio of the deposition values for wet samples to dry pulverized samples was close to unity for most radionuclides. The homogenization of the wet samples is not as efficient in mixing the infrequent particles enriched in refractory nuclides, resulting in a consistent bias toward higher wet values (Krey et al., 1986).

2.4.4.4
PROCESSING EQUIPMENT

The following equipment are used at EML to process soil:

1) Scale - capacity of 50 kg;

2) Sieves - various screen sizes;

3) Splitter - sample reducer, Humbaldt Mfg. Co., Norridge, IL 60656;

4) Crusher - jaw crusher, Morse Bros. Machinery Co., Denver, CO 80214;

5) Blender - twin shell pin intensifier blender, 16 qt capacity, Patterson-Kelly Co., Inc., East Stroudsburg, PA 18301;

6) Pulverizer - pulverizing (hammer) mill with #50 Screen, Weber Brothers & White Metal Works, Inc., Hamilton, MI 49419;

7) Drying oven.

2.4.5 **REPRODUCIBILITY**

The accuracy of the estimated deposit at any site is related to the representativeness of the soil sampling and aliquoting, and to the quality of the analysis. The sampling design should prevent biases and allow errors to be readily determined. A QA program should be maintained at all stages of the project. A detailed discussion of QA for soil sampling is presented in Barth et al. (1989) and van Ee et al. (1990).

2.4.5.1
REPRESENTATIVENESS

If the criteria for selecting a soil sampling site are satisfied, the EML sampling method has been shown to provide reasonable estimates of local and regional fallout. Integrated fallout of ^{137}Cs, ^{90}Sr, and ^{239,240}Pu have been shown to be quite uniform within metropolitan areas characterized by the same annual precipitation. The deposition variability of these radionuclides was <15% (Hardy, 1975). In extensive studies of ^{90}Sr deposition, 50 paired sites, 2-40 km apart, showed an average difference expressed as a percent of the mean of <10% (Hardy and Krey, 1971).

Where it can be assumed that there is little gradient in the cumulative fallout within a city, duplicate soil collection and measurement has inferred a precision of sampling and analysis of ±8% for ^{137}Cs (Beck and Krey, 1980), ±9% for ^{239,240}Pu, ±3% for the ^{240}Pu/^{239}Pu atom ratio, and ±4% for the ^{241}Pu/^{239}Pu atom ratio (Krey and Beck, 1981) for the determination of these values at any given site.

The statistical analyses of common sampling designs, such as random or systematic sampling, are discussed in a familiar text such as Cochran (1977).

In cases where the contaminant may not be uniform in size or concentration, the resulting "hot spots" will be found in some but not all the samples. Uneven physical distribution of particles or particles with a large range of sizes and concentrations complicates the collection of a representative sample and subsampling. Gilbert (1987) discusses different approaches for sampling nonuniformly distributed contamination.

2.4.5.2
ALIQUOTING

Analyses of Utah soils have shown that the prepared soils are sufficiently homogeneous, after following the procedures described above (air-dry, crush, blend, and pulverize), that duplicate aliquots agree within two standard deviations of the counting rate for ^{137}Cs (Beck and Krey, 1980) and the radioassay value for ^{238}Pu, ^{239,240}Pu, and ^{241}Am (Krey and Beck, 1981). A mean deviation of 7% was found for 426 pairs of duplicate soil aliquots, representing the subsampling and analytical errors for ^{90}Sr over an 8-y period (Hardy and Krey, 1971).

2.4.5.3
ANALYTICAL ACCURACY

A. Soil standards.

Two standard reference materials are available through the National Institute of Standards and Technology (NIST, formerly the National Bureau of Standards), SRM 4353 Rocky Flats Soil Number 1 and SRM 4355 Peruvian Soil. SRM 4355 has nonmeasurable radioactivity concentrations for many fallout radionuclides and is intended for use as a blank. Upper limits of the radioactive concentrations are given and can be used to monitor laboratory contamination and background counting rates.

Secondary soil standards, that is, large quantity soil samples that have been dried, blended and pulverized and aliquots of which have been analyzed by inter- and intra-laboratory comparisons, are used at EML as the best possible substitute for a primary standard soil sample for artificial radioactivity. There is no satisfactory way to add a radionuclide to a soil sample so that it represents the chemical and physical form as it exists in the field.

B. Blanks.

Soils collected in 1943, and therefore containing no artificial radionuclides, have served as a quality control blank sample throughout EML's soils programs to monitor contamination by laboratory handling, reagents, and other possible sources.

REFERENCES

Alexander, L. T., E. P. Hardy, and H. C. Hollister
"Radioisotopes in Soils: Particularly with Reference to Strontium-90"
R. S. Caldecott and L. A. Snyder (Editors)
in: Proceedings of a Symposium on Radioisotopes in the Biosphere, University of
  Minnesota, Minneapolis, MN, pp. 3-22 (1960)

Alexander, L. T., R. H. Jordan, R. F. Dever, E. P. Hardy, G. H. Hamada, L. Machta, and
R. J. List
"Strontium-90 on the Earth's Surface "
USAEC Report TID-6567, Office of Technical Information, Washington, D.C., February
  (1961)

ASTM
"Sampling Surface Soils for Radionuclides"
Annual Book of American Society of Testing Materials, C-998-83, Vol. 12.01, p. 792
  (1983a)

ASTM
"Soil Sampling Preparation for the Determination of Radionuclides"
Annual Book of American Society of Testing Materials, C-999-83, Vol. 12.01, p. 796
  (1983b)

Barth, D. S., B. J. Mason, T. H. Starks, and K. W. Brown
"Soil Sampling Quality Assurance User's Guide"
USEPA Report EPA/600/8-89-046, p. 268, March (1989)

Beck, H. L. and P. W. Krey
"Cesium-137 Inventories in Undisturbed Utah Soils - Interim Report on Radionuclides in
  Soils of Populated Areas"
USDOE Report EML-375, June (1980)

Bernhardt, D. E.
"Evaluation of Sample Collection and Analysis Techniques for Environmental
  Plutonium"
USEPA Report ORP/LV-76-5, Las Vegas, NV (1976)

Cochran, W. G.
Sampling Techniques
John Wiley & Sons, Inc. (1977)

Eisenbud, M.
Environmental Radioactivity
Academic Press, Inc., New York, Third Edition, pp. 84-104 (1987)

EPA
"Sample Collection Manual: Guidelines for Collecting Field Samples"
Environmental Protection Agency, Office of Pesticides Programs, Washington, DC,
  March (1979)

EPA
"Preparation of Soil Sampling Protocol: Techniques and Strategies"
Environmental Protection Agency, Office of Research and Development, Report 600/
4-83-020, Washington, DC, August (1983)

Fowler, E. B., R. O. Gilbert, and E. H. Essington
"Sampling of Soils for Radioactivity: Philosophy, Experience and Results"
in: Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants
ERDA Symposium Series 38, CONF-740921, pp. 709-727 (1974)

Gilbert, R. O.
Statistical Methods for Environmental Pollution Monitoring
Van Nostrand Reinhold Company, New York (1987)

Hardy, E. P.
"Depth Distribution of Global Fallout ^{90}Sr, ^{137}Cs, and ^{239,240}Pu in Sandy Loam Soil"
USAEC Report HASL-286, pp. 2-10, October (1974)

Hardy, E. P.
"Regional Uniformity of Cumulative Radionuclide Fallout"
USAEC Report HASL-288, pp. 2-9, January (1975)

Hardy, E. P.
"Plutonium in Soil Northeast of the Nevada Test Site"
USERDA Report HASL-306, pp. 51-85 (1976)

Hardy, E. P. and B. Bennett
"Radionuclide Uptake by Cultivated Crops"
USERDA Report HASL-321 pp. 19-38 (1977)

Hardy, E. P. and P. W. Krey
"Determining the Accumulated Deposit of Radionuclides by Soil Sampling and Analysis"
Los Alamos Scientific Laboratory Report LA-4756, pp. 37-42, August (1971)

Hardy, E. P., P. W. Krey, and H. L. Volchok
"Global Inventory and Distribution of Fallout Plutonium"
Nature, 241, 444-445 (1973)

Hardy, E. P., H. L. Volchok, and P. W. Krey
"Strontium-90 Fallout in Utah"
USAEC Report HASL-257, pp. 71-94, July (1972)

Hardy, E. P., M. Meyers, J. S. Allen, and L. T. Alexander
"Strontium-90 on the Earth's Surface"
Nature, 219, 584-587 (1968)

Healy, J. W.
"Review of Resuspension Models"
W. C. Hanson (Editor)
in: Transuranics Elements in the Environment
USDOE Technical Information Center Report DOE/TIC-22800, pp. 209-235 (1980)

Krey, P. W.
"Remote Plutonium Contamination and Total Inventories from Rocky Flats"
Health Phys., 30, 209-214 (1976)

Krey, P. W. and H. L. Beck
"The Distribution Throughout Utah of ^{137}Cs and $^{239+240}$Pu From Nevada Test Site
 Detonations"
USDOE Report EML-400, November (1981)

Krey, P. W. and C. S. Klusek
"Soil Sampling in Complex Terrain"
J. Radioanalytical Nuc. Chem., 197, 79-98 (1995)

Krey, P. W., E. P. Hardy, and L. E. Toonkel
"The Distribution of Plutonium and Americium with Depth in Soil at Rocky Flats"
USERDA Report HASL-318, April (1977)

Krey, P. W., E. P. Hardy, C. Pachucki, F. Rourke, J. Coluzza, and W. K. Benson
"Mass Isotopic Composition of Global Fall-Out Plutonium in Soil"
in: Transuranium Nuclides in the Environment, IAEA-SM-199/39, pp. 671- 678 (1976a)

Krey, P. W., E. P. Hardy, H. L. Volchok, L. E. Toonkel, R. H. Knuth, R. Coppes, and
  T. Tamura
"Plutonium and Americium Contamination in Rocky Flats Soils, 1973"
USAEC Report HASL-304, March (1976b)

Krey, P. W., C. S. Klusek, C. G. Sanderson, K. M. Miller, and I. K. Helfer
"Radiochemical Characterization of Chernobyl Fallout in Europe"
H. L. Volchok and N. Chieco (Editors)
in: A Compendium of the Environmental Measurements Laboratory's Research Projects
  Related to the Chernobyl Nuclear Accident,
USDOE Report EML-460, pp. 155-213, October (1986)

Russell, R. S., B. O. Bartlett, and R. S. Bruce
"Appraisal of the Long-Term Nuclide Contamination Hazard"
D. W. Bensen and A. H. Sparrow (Editors)
in: Survival of Food Crops and Livestock in the Event of a Nuclear War
AEC Symp. Ser. 24 (CONF-700909), Natl. Tech. Info. Service, U.S. Dept. of Commerce,
Springfield, VA, pp. 548-565 (1971)

Schulz, R. K.
"Soil Chemistry of Radionuclides"
Health Phys., 11, 1317-1324 (1965)

Sehmel, G. A.
"Particle Resuspension: A Review"
Environment International, 4, 107-127 (1980)

van Ee, J. J., L. J. Blume, and T. H. Starks
"A Rationale for the Assessment of Errors in the Sampling of Soils"
USEPA Report EPA/600/4-90/013, p. 57, May (1990)
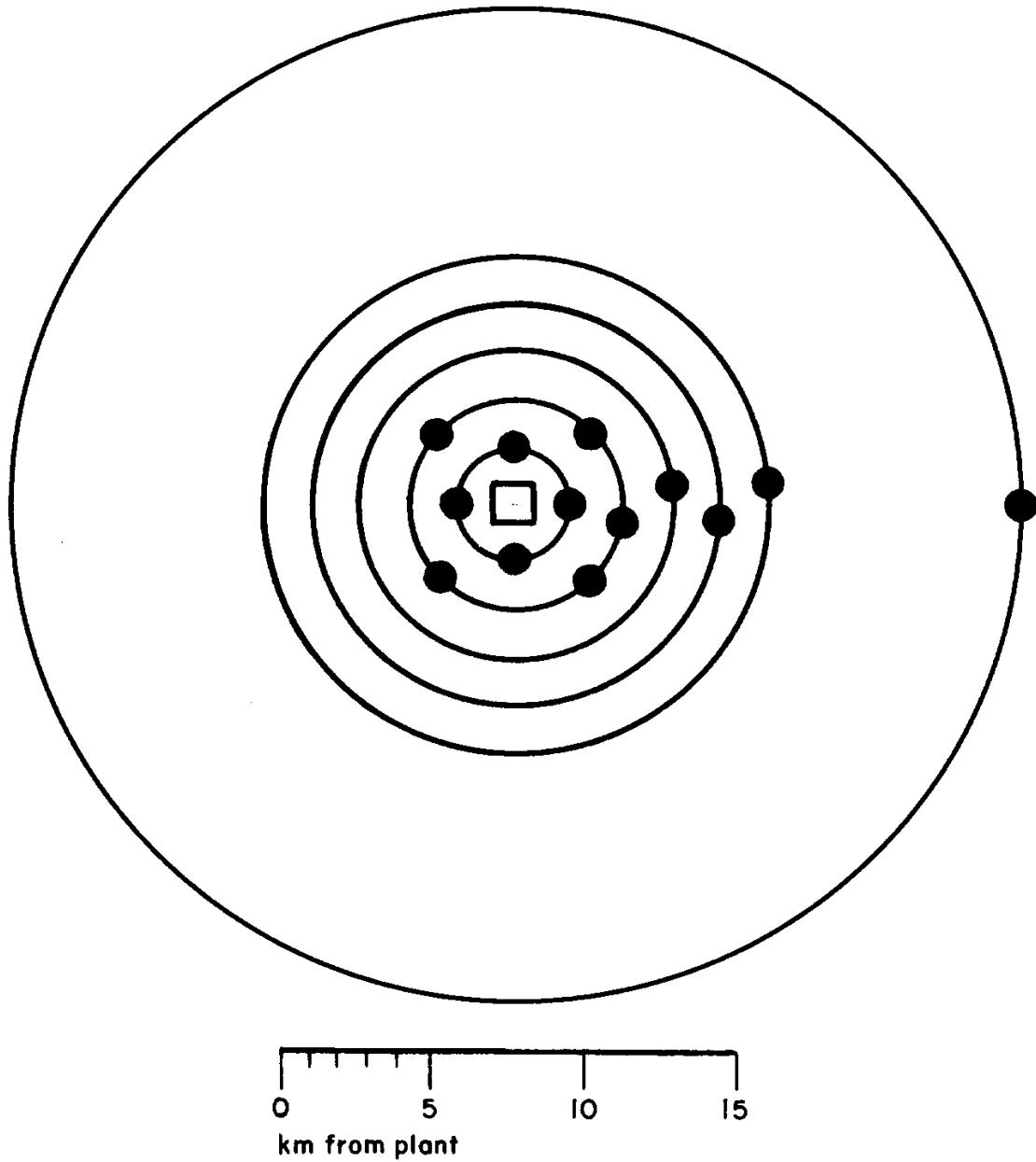
Figure 2.10.   A suggested distribution of sampling sites covering the area surrounding a
                     plant, with emphasis on the downwind direction.

2.5 SEDIMENT

Contact Person : Mathew A. Monetti

2.5.1 SCOPE

Discussed in this section are the methodologies used at EML for obtaining sediment cores from inland lakes and reservoirs for reconstructing the deposition histories of energy-related pollutants. Topics included are the relationship of lake bathymetry to core quality, a general description of the operation of the corer from our specially designed catamaran, and the procedures for extruding the cores. The catamaran itself is discussed in Section 6.7.

2.5.2 INTRODUCTION

Lake and reservoir sediments offer unique substrates for investigating the occurrence of many energy-related pollutants since they are the main "sink" for materials entering watersheds and may be dated by radioactive methods so as to provide a depositional history (MARC, 1985). Furthermore, accurate coring, e.g., minimal disturbance of sediments, is of great importance to paleolimnological investigations, especially those aimed at reconstructing the deposition history and/or inventories of pollutants deposited through atmospheric processes. Errors caused by a poorly performing coring system or by taking sediment cores from locations in lakes that are not representative of atmospheric deposition (e.g., areas affected by sediment focusing or excessive erosion) can often lead to an erroneous interpretation of actual events (Heit and Miller, 1987).

To properly assess geochronologies of sediments in a meaningful way, methodologies must be available for taking large volume, undisturbed sediment cores, so that multiple parameter analyses can be performed at critical core depths. As stated in Burke (1968), for the commonly used 4-7 cm diameter sediment corers, increased sample size can be obtained only by homogenization of longer segments of core, with consequent blurring of stratigraphic details. For isotopic dating procedures, the demand for really large samples (>200 g wet weight) presents an even more severe problem. Only by increasing the area of the column sampled can one improve the sensitivity in the study of sediment horizons.

Unfortunately, most larger area coring devices are of such size and weight that they can only be used on oceanographic-sized vessels, making them unfit for studies of most lakes and reservoirs.

As further stated in Burke (1968), the study of vertical profiles of the concentrations of fallout radionuclides (which have been available for sedimentation only since the early 1950s in most places) also demands efficient sampling of the topmost layers of sediments and their recovery in situ. Some evidence from studies of more conventionally collected cores indicates that these upper layers are commonly lost, either by washout at the top of the core tube or by being swept away from the point of impact by the shock wave of rapidly falling corers (Sachs and Raymond, 1965). Also, as stated in McIntyre (1971), a peculiarity of many gravity corers is that the length of the core retrieved may be considerably less than the penetration depth of the core barrel into the sediment. This is likely to cause a vertical redistribution of the sediment and hence an erroneous chronology. Also core tubes having diameters <10 cm are subject to loss of surface sediment (McIntyre, 1971).

2.5.3 **EQUIPMENT DESIGN AND APPLICATION**

We have developed a large diameter (21 cm), slow penetration sediment corer, and a 27 ft (7 m) transportable research vessel ("Sedimental Journey") to avoid the pitfalls described above. This equipment, whose design and construction are described in detail in Section 6.7, has allowed us to take large volume, undisturbed cores from many locations in the U.S. and measure increments of sediment as small as 1 cm for a suite of toxic trace substances and environmental tracers indicative of both natural and anthropogenic origins. In fact, sediment cores taken with the tripod-sphincter corer have been shown to be comparable in quality to those taken with the much larger "Soutar type" box corers which, because of their size, can only be used successfully on oceanographic vessels (Burke et al., 1983).

Examples of highly resolved sediment cores that can be obtained with our sediment sampling equipment are shown in Figures 2.11-2.13. Plotted in these figures is the ^{137}Cs activity per unit area (Bq m^{-2}) versus depth (cm) for sediment cores taken from Cayuga Lake, Ithaca, NY (Figure 2.11), and Deer Creek Reservoir, Provo, UT (Figure 2.12). Clearly, the ^{137}Cs distributions closely follow the historic deposition

pattern of ^{137}Cs from atmospheric weapons testing shown in Figure 2.13. The two easily distinguished ^{137}Cs peaks follow the major cycles of fallout from atmospheric weapons testing in 1959 and 1963. Furthermore, there is a rapid drop of ^{137}Cs activity in sediments younger than 1963 following the Nuclear Test Ban Treaty between the U.S. and U.S.S.R. Lastly, the activity after the onset of atmospheric weapons testing follows closely that of historic fallout patterns with little debris diffused or mixed below the depth corresponding to 1951.

Further confirmation of our ability to take quality cores with this system is shown by the use of the SNAP-9A (Systems for Nuclear Auxiliary Power) Pu isotope signature. In 1964, the U.S. satellite, SNAP-9A, which used ^{238}Pu as a heat source for generating power, disintegrated after entering the atmosphere of the Southern Hemisphere. As a result, there was a sharp rise in the stratospheric concentration of ^{238}Pu relative to $^{239+240}$Pu in 1964 in the Southern Hemisphere. The pulse of ^{238}Pu from SNAP-9A did not reach the surface of the Northern Hemisphere until 1966, at which time a distinct rise in the ratio of ^{238}Pu to $^{239+240}$Pu activity also occurred. It is the increase in this ratio which is used as a geochronological marker for establishing the year 1966 in a sediment core. This is clearly shown in Figure 2.14 where the ^{238}Pu/$^{239+240}$Pu activity ratio increases between 13 and 11.5 cm deep in the sediment core from Cayuga Lake, NY.

The remainder of this section is a description of the procedures that are used by EML to take sediment cores from inland lakes and reservoirs using the Sedimental Journey and tripod corer.

2.5.4 PROCEDURES

2.5.4.1
GENERAL OPERATION

Procedures for the preparation and operation of the Sedimental Journey are described in detail in Section 6.7. The basic coring strategy is to launch the boat and determine the best sampling sites through a bathymetric survey of the lake or reservoir, obtain the

cores, extrude the cores immediately after their retrieval, and, finally, remove the boat from the water with the trailer.

<div align="center">

2.5.4.2
BATHYMETRIC SURVEY

</div>

Bathymetric surveys are always conducted prior to coring since such data are usually not available. It has been our experience that even when bathymetry is available, it is usually inaccurate or out of date and hence misleading. This can be a severe problem because the lack of accurate bathymetric data will almost certainly result in a failure to locate suitable coring locations within a lake or reservoir.

In general, it has been our aim to take replicate cores from those portions of lake basins which are the least likely to be affected by processes known to disturb sediments, such as, excess terrestrial runoff (erosion), sediment slumping, sediment resuspension, and sediment focusing (Davis and Ford, 1982; Edgington and Robbins, 1977; Heit and Miller, 1987). In this regard, the bathymetric survey is used to select sites located in relatively large, hypolimnetic basins, as far away as possible from shoreline areas prone to erosion. Inlet and outlet areas, as well as locations at the confluence of rivers or close to dams are also avoided.

For our bathymetric survey work we use a Lowrance graph recorder sonar unit. This device is capable of accurately graphing a lake bottom to a depth of 300 m (900 ft). Our location is determined using a micrologic explorer Loran system.

<div align="center">

2.5.4.3
SEDIMENT SAMPLING

</div>

All of the sediment cores are taken with a sphincter corer with a tripod modification capable of taking a 21 cm diameter, 90 cm long core. The design and mechanics of the corer are described in detail in Section 6.7.

The device uses the barrel and nose-cone of the original 21-cm sphincter corer (Burke, 1968), but is mounted in a tripod (see Section 6.7.4). The operation of the sphincter is not changed by this design, the tripod frame only provides stability to keep the corer in a vertical position while it penetrates. This design also allows the corer to be lowered to the bottom very slowly, with minimal sediment disturbance. The core barrel is driven into the sediment only by the force of weights mounted on the weight stand (see Section 6.7.4). Varying the amount of this weight gives control over penetration into various sediment types. It should be noted that the tripod corer is "manageable" on small boats and is easily disassembled for transporting or shipping.

The corer is lowered at 100 m min^{-1} (75 m min^{-1} in fairly active weather) until it is 5 m from the bottom (observable with the Sonar unit). It is then slowed to 10 cm min^{-1}, or as slow as the winch will operate, for placement on the bottom. The feet of the tripod contact the sediment first. The barrel continues to descend, penetrating the sediment by the force of the weights contained in the weight stand. The core barrel stops its downward travel when the weight stand comes in contact with the guide ring (see Section 6.7.4.2), or when the resistance of the sediment is sufficient to stop the downward penetration. When the weight stand comes to a stop, the weight is taken off the spring-loaded release pin and the pin retracts (see Section 6.7.4). This should be completed within 60 sec after the corer reaches the bottom. The pull on the wire to retrieve the corer begins immediately and closes the diaphragms of the core catcher and the top valve. Stress is taken off the closure wires by a stop which engages the bottom of the release mechanism where force is exerted during retrieval.

The corer is pulled out of the sediment at the slowest winch speed, or about 10 cm min^{-1}. In the past, pullout forces have not exceeded 4450 N (1000 lb force) and are generally somewhat lower. After pullout the corer can be retrieved at a rate of 50 m min^{-1}.

2.5.4.4
SEDIMENT EXTRUSION

More than one method has been employed to extrude sediments from the core barrels since EML started coring in the early 1970s. In all of these methods, the sediments are always extruded immediately after retrieval. Water retained above the core is siphoned or pipetted off and may be reserved for analyses. In most cases, contiguous 1 cm increments

are taken from the top of the core to ~ 40 cm, and 2 cm sections are taken thereafter. The outer ~ 1 cm of each layer of sediment is always discarded, resulting in ~ 19 cm diameter sections. We have found that if this outer ring of sediment is not eliminated, contamination may result from surface sediments "dragging down" to subsurface depths via friction with the core barrel wall or through other physical processes. Lastly, glassware and covers used for sample storage are prewashed with 1:1 HNO_3, rinsed with double-deionized water, followed by acetone and methyl-alcohol washes. All of the glassware is dried and sealed under laminar flow clean stations until the time of use. Precleaned glass jars are used, which can be obtained from several suppliers.

Prior to extrusion, the length of the core and its distance from the top of the barrel are measured and noted. After the excess water is removed by siphon or pipette, the sediment is then carefully removed in 1 cm increments with a large Teflon-coated spoon bent into an "L" shape. The accuracy of this procedure is ensured by placing the sediment in precalibrated 16-oz (500 mL) wide-mouth glass jars, and noting the actual extrusion depth relative to the initial location of the top of the core as measured immediately after core retrieval. In addition, the core barrels are marked at 5 cm increments for further calibration; thus, errors in extrusion are identified.

Once the surface sediment is removed, the remaining sediment is usually firm and is not easily disturbed. The rest of the core can be sectioned in the following manner. A stainless steel piston is fitted inside the top of the core barrel and contacts the sediment. While this piston is held in place, the core is laid down on its side, the bottom core catcher is removed, and another piston is placed into the bottom of the core. The core is then listed upright again and placed on an extruding stand. Next, the top piston is removed and the bottom one is pushed up through the core barrel until the sediment becomes flush with the top of the barrel. The remainder of the sediment is extruded at 1 to 2 cm intervals by pulling the barrel down to designated marks on the attached scale. The outer ring is removed from the protruding sediment; the rest of the interval is sliced and packed into sampling jars. Sediment is sectioned in this way to a desired depth or to the bottom of the core. A description of the construction and design of the extruder used to section the core is given in Section 6.7.4.

The reproducibility of this technique is demonstrated in Figure 2.15, which compares the ^{137}Cs distribution in two sediment cores (A and B), taken from separate but nearby locations (within 0.5 km) in Cayuga Lake, Ithaca, NY. Although the sedimentation rate

was slightly different for the two locations (Heit et al. 1986), as would be expected from bathymetric variations within a lake basin, even for nearby locations, the ^{137}Cs distribution patterns were remarkably similar with only small differences occurring among the major peaks which represent fallout from atmospheric weapons testing in 1959 and 1963. This agreement ($\pm$ 2 cm) between the patterns of ^{137}Cs distributed in these independent cores attests to the reproducibility of our current method of sediment extrusion.

REFERENCES

Burke, J. C.
"A Sediment Coring Device of 21-cm Diameter with Sphincter Core Retainer"
Limnology and Oceanography, 13, 714-718 (1968)

Burke, J. C., R. E. Hamblin, and S. A. Casso
"Tripod Modification Of Sphincter Corer: Construction, Operation, Core Extrusion and Sampling Efficiency"
Woods Hole Oceanographic Institution Technical Report WHOI-83-36, Woods Hole, MA 02543, pp. 1-13 (1983)

Davis, M. B. and M. S. Ford
"Sediment Focusing In Mirror lake, New Hampshire"
Limnology and Oceanography, 27, 137-150 (1982)

Edgington, D. N. and J. A. Robbins
"Patterns Of Deposition of Natural and Fallout Radionuclides to Limnological Processes"
J. O. Nriagu (Editor)
in: *Environmental Biogeochemistry - Metals Transfer and Ecological Mass Balances*
Ann Arbor Science, Ann Arbor, MI, pp. 705-729, Vol. 2 (1977)

Heit, M. and K. M. Miller
"^{137}Cs Sediment Depth Profiles And Inventories In Adirondack Lake Sediments"
Water, Air and Soil Pollution, 3, 243-265 (1987)

Heit, M., Y. L. Tan, K. M. Miller, J. Quanci, C. Marinetti, and S. Silvestri
"The Sediment Chronology and Polycyclic Aromatic Hydrocarbon Concentrations and
  Fluxes in Cayuga Lake, NY"
U.S. Department of Energy Report EML-451 (1986)

MARC: Monitoring and Assessment Research Centre
"Historical Monitoring"
MARC Report 31, University of London, London, England (1985)

McIntyre, A. D.
"Efficiency of Benthos Sampling Gear"
N. A. Holme and A. D. McIntyre (Editors)
in: *Methods for the Study of Marine Benthos*
International Biological Program Handbook No. 16, Blackwell Scientific Publications,
  pp. 140-146 (1971)

Sachs, P. L. and S. O. Raymond
"A New Unattached Sediment Sampler"
J. Marine Res., 23, 44-53 (1965)
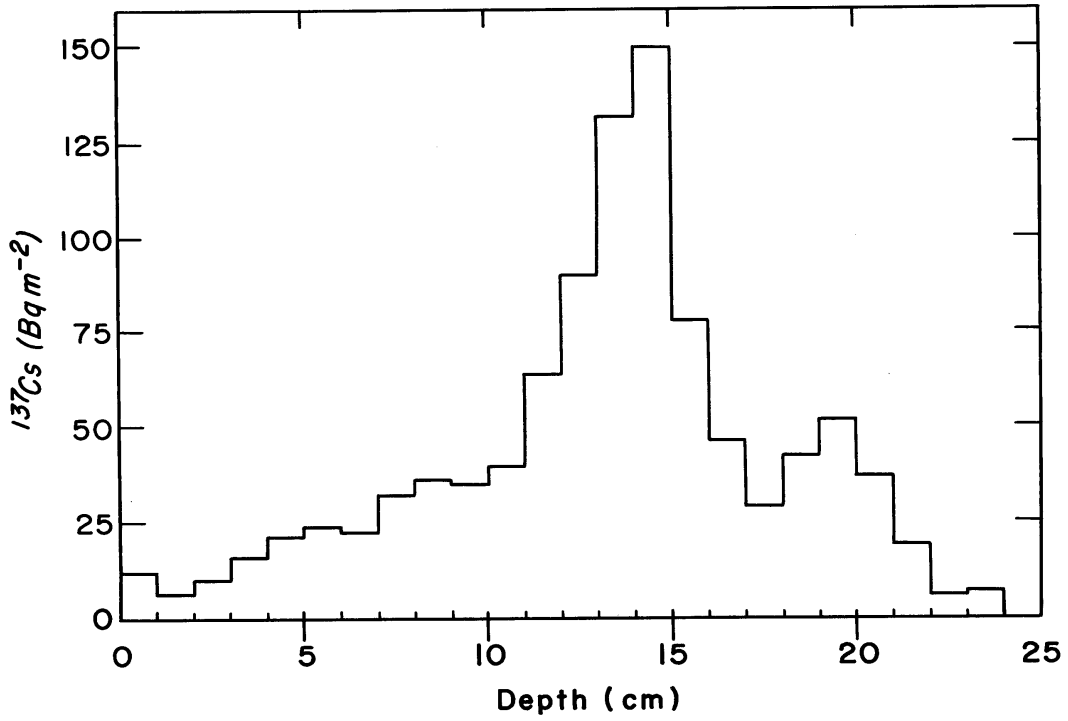
Figure 2.11. ^{137}Cs activity per unit area versus sediment depth for Cayuga Lake,
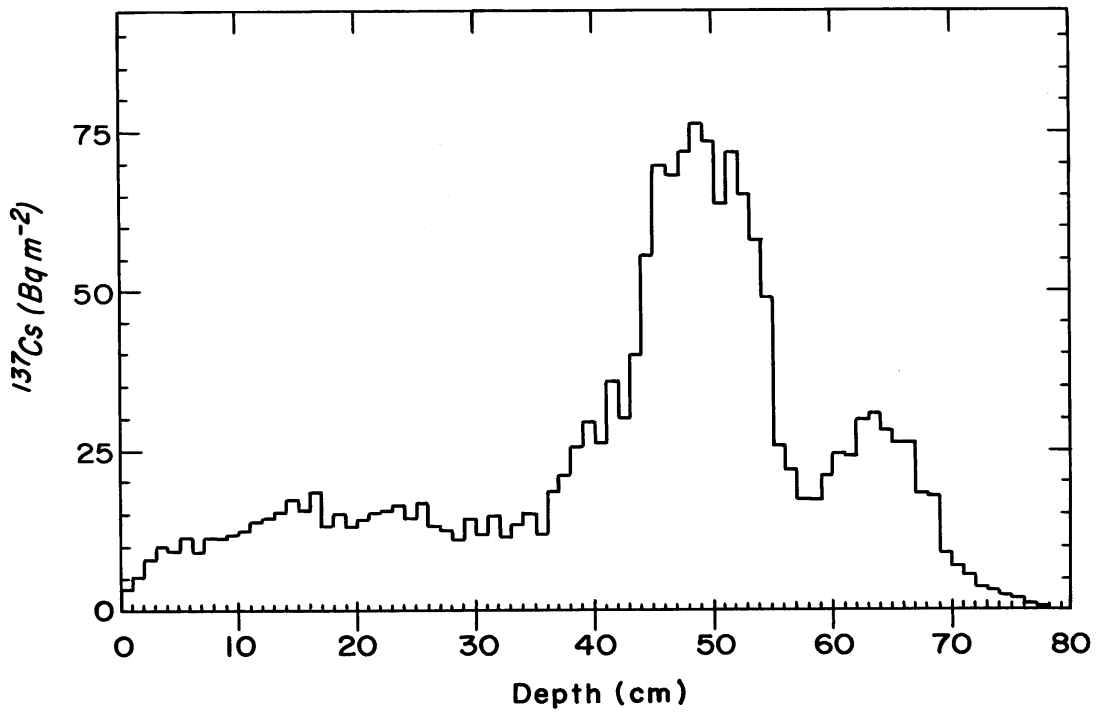New York.

Figure 2.12. ^{137}Cs activity per unit area versus sediment depth for Deer Creek
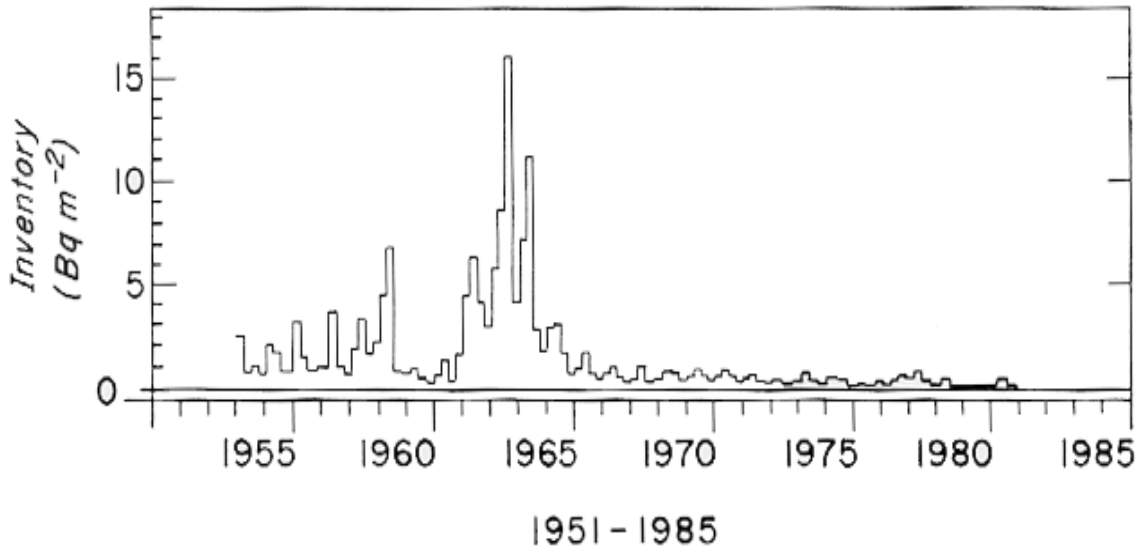Reservoir, Provo, Utah.

Figure 2.13. Historic deposition pattern of ^{137}Cs in the 40° - 50° latitude band from atmospheric weapons testing.
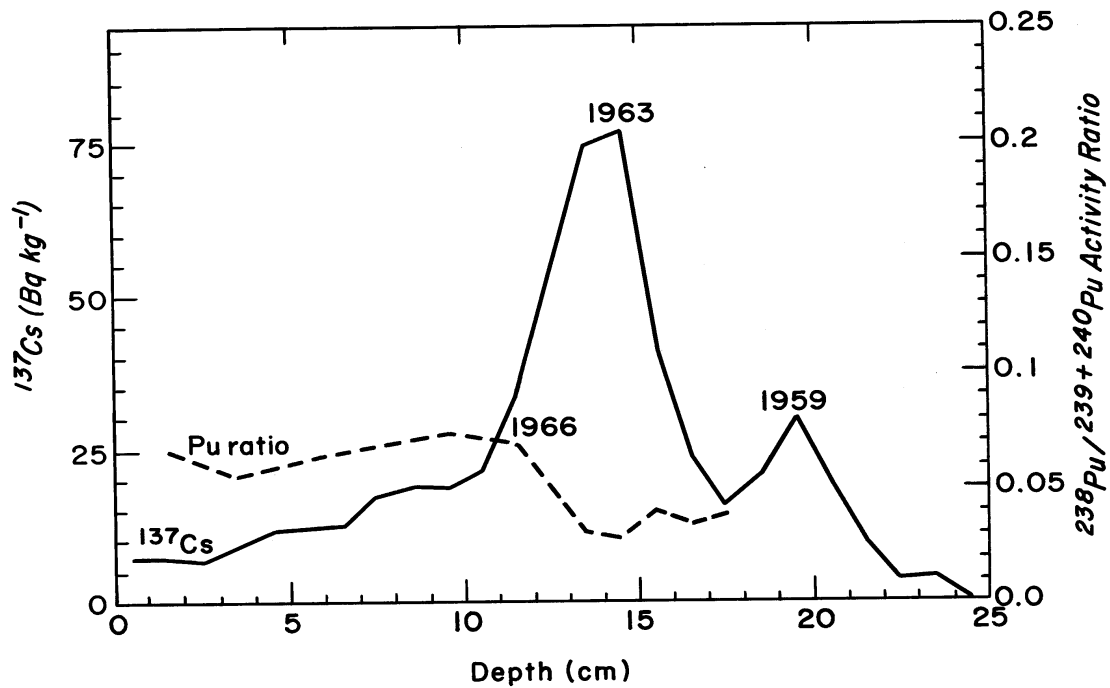

Figure 2.14. ^{238}Pu/$^{239+240}$Pu activity ratio in relationship to ^{137}Cs concentration in the sediments of Cayuga Lake, New York.
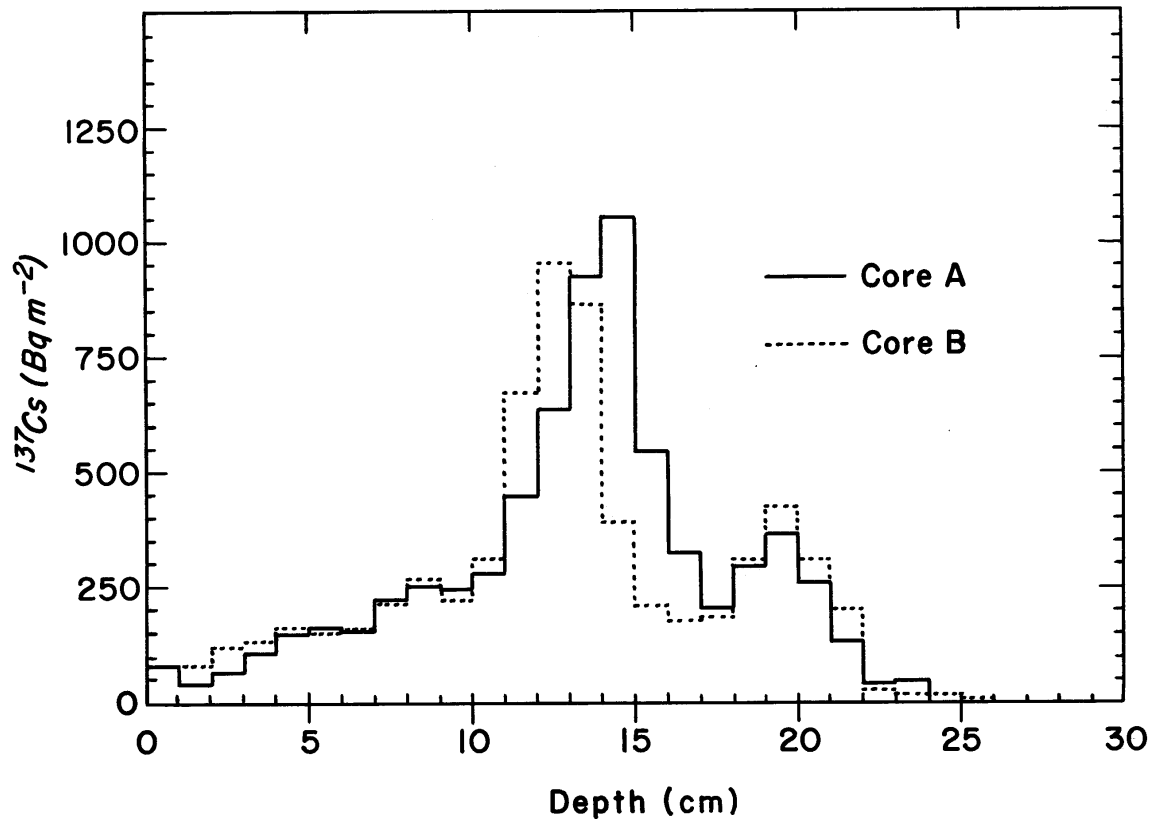
Figure 2.15. ^{137}Cs distribution in two sediment cores taken from nearby locations in Cayuga Lake, New York.

2.6 URANIUM AND RADIUM IN WATER

Contact Person(s) : Isabel M. Fisenne

2.6.1 SCOPE

A simple method for the field collection of uranium and radium in 10 L of fresh water (tap, well, river) is described. Similar collection methods for radium are in use elsewhere, notably at the New York University Institute of Environmental Medicine.

This method has been used to collect uranium and radium from fresh water samples at environmental levels. The use of the ion exchange collector concentrates the uranium and radium in the water samples and permits shipment of the samples within national and international regulations. Water samples collected with this system have been sent to EML from locations within the continental U.S., the middle East, and Africa.

2.6.2 APPARATUS

The ion exchange collector is a modified version of the fallout collector described in Section 2.3.3. This unit consists of a funnel and ion exchange column constructed of polyethylene. The funnel is welded to a threaded cap which is attached to the top of the ion exchange column. The bottom of the column is threaded for a tapered fitting, which in turn has a small cap at the end. The funnel and the tapered fitting are replaced with standard bottle caps for return shipment to EML.

2.6.3 PROCEDURE

The column is packed with a plug of glass wool, a 1 cm plug of Whatman No. 41 paper strip (Note 1), 150 mL of mixed anionic-cationic resin (Note 2), and a top plug of filter paper strip. The threaded tapered fitting (outlet end of the column) is taped to the column to assure leak-proof operation during sampling.

The column is attached to a funnel marked for a 10 L volume, it is then placed in a stand and 10 L of the water to be sampled is poured into the funnel. The bottom cap is removed from the tapered fitting, allowing gravity flow of the water. The collection time is about 3 h.

Upon return to EML, the resin and paper pulp are pushed out of the column body into a 250-mL platinum crucible. The glass wool is discarded. The resin may be analyzed sequentially for isotopic uranium and radium (see Procedure Se-01, Section 4).

Notes:

1. Filter paper pulp must not be used, as the flow rate through the column is too slow. Narrow strips cut from filter discs are recommended.

2. The resin is an equal mixture of Bio-Rad AG1-X4 in the H^+ form (20-50 mesh) and Bio-Rad AG 50-X8 (20-50 mesh).

2.7 **FOOD**

(see Volume II)