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RADON FLUX MEASUREMENT AND COMPUTATIONAL METHODOLOGIES

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ABSTRACT

A detailed review has been performed of present technology for radon flux measurement and calculation methods. The review concentrates on technology which was utilized under the Uranium Mill Tailings Remedial Action (UMTRA) Project and which is available for future remedial action design applications. Five main radon flux measurement methods are evaluated: the accumulator can, a small charcoal sampler, a large-area charcoal sampler, the "Big Louie" charcoal sampler, and the charcoal tent sampler. These sampling methods are evaluated with respect to their theoretical maximum sampling times, and equations are given to allow predictions of maximum sampling times if any of these samplers are modified.

An experimental comparison of the five flux measurement techniques was conducted using the thin reference radon flux source at the Bendix Technical Measurements Center (TMC). Based on gamma-ray analysis and alpha counting by Rogers and Associates (RAE), excellent agreement was obtained between the measured radon fluxes and fluxes predicted from radium and emanation measurements by the TMC. The respective mean radon fluxes, standard deviations, and numbers of measurements (in parentheses) for each of the five measurement methods were: 29.6 ± 3.5(9); 32.4 ± 2.9(12); 30.2 ± 2.7(6); 29.5(1); and 35.6 ± 5.2(3) pCi $m^{-2}s^{-1}$. The flux estimated from radium and emanation measurements was 30.4 ± 2.1 pCi $m^{-2}s^{-1}$. Fluxes based on gamma-ray analyses by Pacific Northwest Laboratory agreed within 2.5 percent and 3.1 percent of measurements by RAE for the two cases in which comparison analyses on the same charcoal samples were available, indicating excellent agreement in the independent calibrations of these two facilities.

Mathematical methods for predicting radon fluxes are deemed adequate both for remedial action design problems and for more complicated numerical simulations of climatological effects, sampler design problems, and other applications. One-dimensional, steady-state diffusion codes with multi-region capabilities (such as RAECO) are suitable for most applications, and utilize rapid analytical solutions to the diffusion equations. Finite-difference methods are required for most cases of greater complexity, and these have been developed for various time-dependent cases of 1-, 2-, and 3-dimensional diffusion.

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1.0 INTRODUCTION AND BACKGROUND

The U.S. Department of Energy (DOE) is conducting several remedial action programs involving radium and uranium contamination. Radon gas releases are generally associated with radium; thus, measurement and calculation of radon fluxes is necessary in some of these remedial action programs. The Uranium Mill Tailings Remedial Action (UMTRA) Project, managed by the Albuquerque Operations Office, DOE, Albuquerque, New Mexico, supported research and development on radon flux measurement and calculation in connection with research on tailings cover design, and also to develop the measurement technology in case radon flux measurements were required for the specific remedial action projects. Although flux measurements will not be made in support of specific site remedial actions, this report has been prepared to summarize the progress to date for application by other interested groups. Radon flux measurements are still important because a radon flux standard is part of the Environmental Protection Agency's (EPA) criteria for the UMTRA Project. Radon flux is also an important component in the design of the remedial actions. The DOE has established a Technical Measurements Center (TMC), operated by Bendix Field Engineering Corporation, to establish standardized procedures for radiometric determinations for all of the DOE Remedial Action Programs. The study reported herein is a review of experimental and calculational methods for determining radon flux as used in the research aspects of the UMTRA project. This review is provided to the TMC for documentation of these methods and assessment of the state-of-the-art in radon flux estimation.

1.1 PURPOSE OF THIS STUDY

The primary objective of this project is to critically review, evaluate and document the present status and state-of-the-art methodologies that have been used in the UMTRA Project for the quantitative determination of the radon flux by:

- Experimental measurements.
- Analytical and numerical evaluations and predictions.

Furthermore, the thin source radon flux facility at the TMC was used to intercompare the principal methods for radon flux measurement used in UMTRA research. The methods and results of this intercomparison are summarized in this report.

This review is directed at providing the technical bases needed to select methods and prepare standardized procedures for the quantitative determination of radon fluxes.

1.2 SIGNIFICANCE OF RADON IN THE ENVIRONMENT

Uranium-238, which has a mean radioactive lifetime of 6.4 billion years and is distributed throughout the earth's crust at about 3 ppm, (1 pCi/g), supports a series of 18 radioactive decay products (Ro81a), as shown in Figure 1.1. Only one of these radioactive decay daughter products, viz., radon-222 or Rn-222 and commonly referred to simply as "radon", is a gas at ambient temperatures and pressures. There are two other naturally occuring radon isotopes; radon-220 (called thoron) which emanates from the decay of thorium-232 and radon-219 (called actinon) which emanates from the decay of uranium-235. However, thoron and actinon with radioactive half-lives of 55.6 seconds and 3.96 seconds respectively, generally provide only a negligible contribution to the radiological impact arising from naturally occuring radon isotopes because of the much greater half-life (3.82 days) of radon.

Because radon is essentially an inert gas it is able to readily migrate through soil, air and most other materials. A typical flux of radon emanating from terrestrial radium sources and migrating into the atmosphere is about 1 pCi per square meter per second. This continuous input to the atmosphere primarily from the lithosphere results in an average outdoor radon concentration of about 0.2 pCi per liter of air.

Although radon itself is only a slight radiological hazard, this highly mobile, ubiquitous gas serves to broadly disperse its succeeding short-lived, alpha-emitting daughter nuclides which are significant contributors to the radiological exposure of humans. The radiologically significant, moderate-lived decay products of radon depicted in Figure 1.1, viz., Po-218 (RaA), Pb-214 (RaB), Bi-214 (RaC), and Po-214 (RaC') are chemically and physically active species, and their life cycle in the environment can be varied and complex.

During the milling of uranium ore, the ore material is crushed to facilitate processing and only a negligible fraction of radium, the radioactive precursor of radon, is removed. Because uranium and radium in uranium ores are present at concentrations typically several hundred times above those in normal soils, radon fluxes are released to the environment from the resulting mill tailings with a similar degree of elevation above the fluxes from natural soils (Be75, Mar77, NRC80).

The major potential human radiation exposure pathways resulting from uranium tailings impoundments are shown schematically in Figure 1.2. Radon releases are generally the dominant potential public health hazard (Pe67) because of the nature of the radon transport processes that can occur.

1.3 ORGANIZATION OF THIS REPORT

Section 1.0 of this report provides an introduction to the significance of radon in the environment. The purpose of this study and its relationship to the UMTRA Project is also stated.

Section 2.0 reviews the common methodologies and equipment employed in the experimental measurement of radon fluxes. The significance of spatial and temporal variations in radon fluxes from typical sites is considered. The requirements for accommodating these variations for an acceptable experimental assessment of a site are quantitatively addressed. Sources of errors and uncertainty for the experimental methodologies are then evaluated.

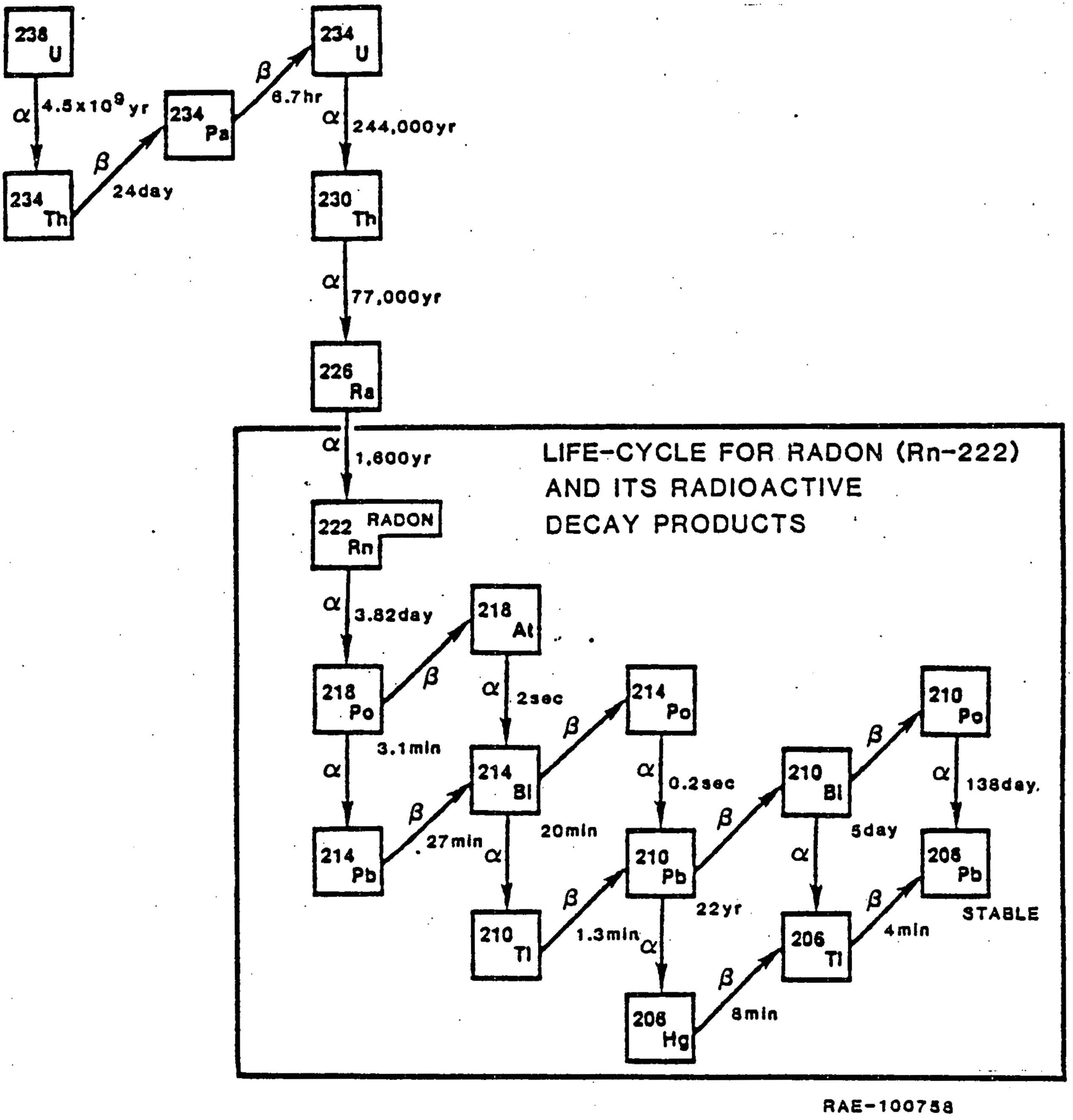


FIGURE 1.1. RADIOACTIVE DECAY SERIES WHICH PRODUCES RADON (Rn-222) AND ITS RADIOACTIVE DECAY PRODUCTS.

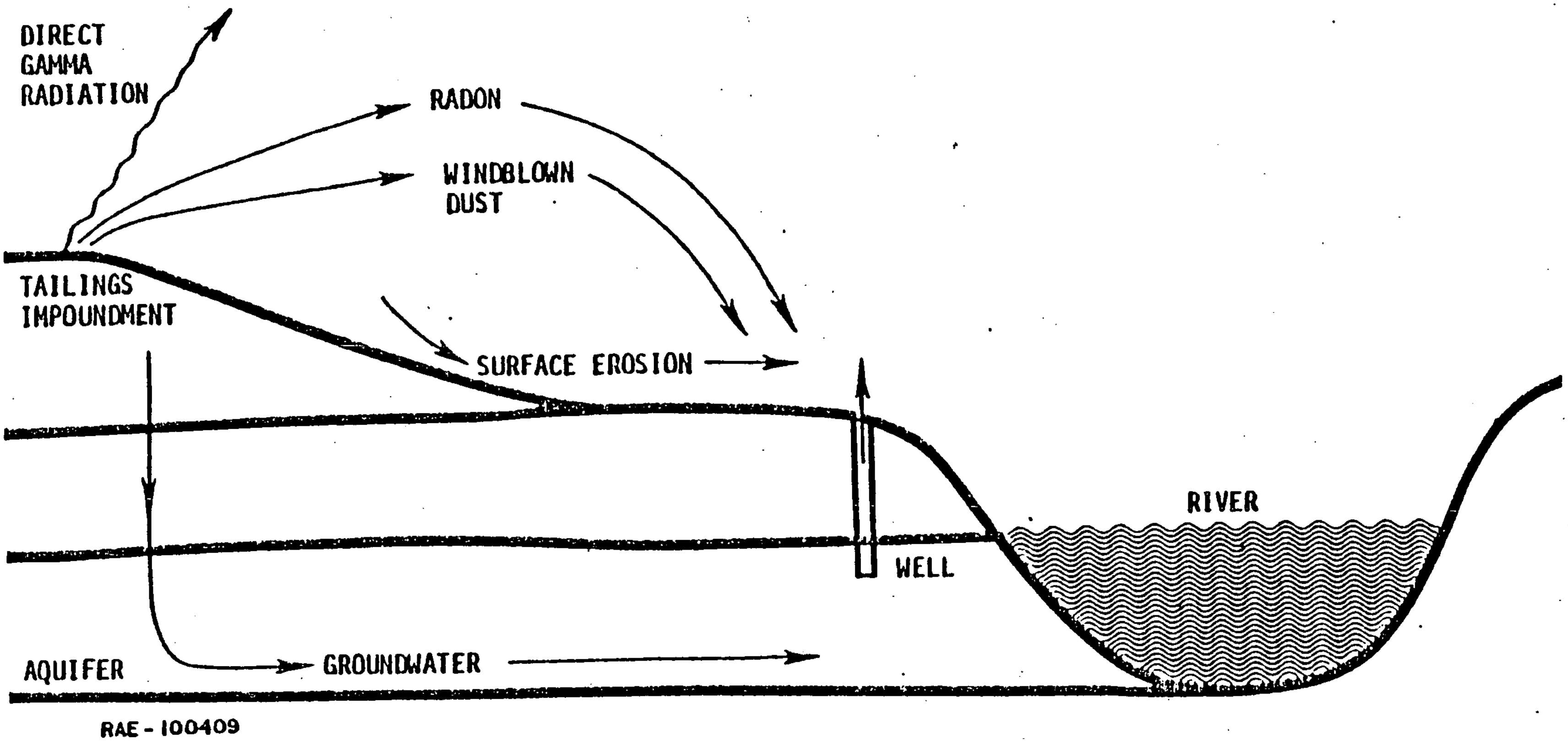


FIGURE 1.2. MAJOR HUMAN RADIATION EXPOSURE PATHWAYS FROM URANIUM TAILINGS.

Section 3.0 addresses the use of theoretical and computational models for the estimation and evaluation of radon fluxes. The physical transport processes that lead to radon migration are defined and the standard radon transport equation is given together with typical analytical solutions for simple migration models. A review is also made of the qualified computer codes that can be employed to evaluate radon fluxes for various disposal site models. A comparison is made of the applications of these codes.

Section 4.0 contains the results of flux intercomparison experiments conducted at the thin reference flux facility at the Bendix Laboratories in Grand Junction, Colorado.

Section 5.0 provides a brief summary and discussion of the major methodologies considered in this review.

An extensive Bibliography is provided of the major published literature related to radon flux measurements, analysis and evaluation.

2.0 EXPERIMENTAL RADON FLUX MEASUREMENTS AND METHODOLOGIES

2.1 BACKGROUND

A variety of experimental methods and techniques have been proposed and used to measure or infer the flux of radon from a body of porous material (Ro82b). These methods and techniques all depend upon the measurement of radioactive decay events arising from the disintegration of radon nuclei or those of its radioactive daughters. Variations in environmental parameters often alter radon fluxes, and sometimes dictate the choice of radon flux measurement methods. Furthermore, the measurement methods and techniques may be currently classified as:

- Discrete sampling measurements of the instantaneous radon flux (such measurements are often referred to generically as "grab" sampling).
- Integrated sampling measurements of the accumulated or summed radon flux over some prescribed time period.
- Continuous sampling measurements of the time varying radon flux over some prescribed time period.

Each of these methods has been employed extensively in environmental radon flux monitoring, and the important experimental techniques and apparatus employed in these three classifications will be reviewed.

2.2 ENVIRONMENTAL CONSIDERATIONS

Of particular importance to experimental methodologies is the observation from many radon flux experiments that time variations of the radon flux from a given source in environmental situations can be significant and complex. For example, if the release of radon to the atmosphere from radium sources within a natural soil is considered, then changes in ambient environmental temperature (Ba69, Ga82), atmospheric pressure (Bat78, Cl74, Du76, Wi72), winds (Du76, Kr64, Is70, Kr64, Pe66), humidity (Str82, Th81), precipitation (rain, snow, ice) (Pe65, Pe66, Cou77), vegetation and soil surface characteristics (cracks, holes, rocks, etc) may perturb the average radon flux from the soil to the atmosphere (Bav56, Mo80, Ro83). These factors can produce variations in the mean radon flux over time periods ranging from minutes, hours and days to years for seasonal and climatic changes (Ba69, Me73). These temporal variations in the radon flux can range from nearly complete suppression of the flux to an order of magnitude or more enhancement. Generally, for regulatory control and monitoring of tailings and tailings covers, what is desired is an accurate estimate of the mean radon flux over a designated time period such as a year.

In addition to temporal variations in the radon flux from a soil surface, there are significant spatial variations which might arise due to inhomogeneities in the cover soil, or in tailings properties such as the radium distribution (Bat78, Bo80, Sa81a). Thus, the possibility for significant spatial and temporal variations in the radon flux must be recognized and evaluated so that satisfactory experimental measurements can be made.

2.3 INSTANTANEOUS RADON FLUX

It is important to recognize that the instantaneous measurement of the radon flux taken at a single location should be used primarily as a benchmark for certain calculations or as a contributory measurement to a data base consisting of many measurements taken at different times and places at the site (Ro82b, You83).

The instantaneous radon flux is usually measured with an accumulator can (Be75, Kr64, Ni81) by measuring the rate of radon concentration increase in the known volume (Mo80). The radon flux can also be estimated by measuring the vertical concentration profile of radon in the atmosphere (Ki70, Wi72). However, this latter experimental method is primarily qualitative and has received only limited use. Therefore, only the accumulator-can method will be considered in detail.

2.3.1 Accumulator

To measure the radon flux using the accumulator method, an open-ended container is sealed to the soil surface either by pressing it gently into the surface or by sealing the container-soil boundary with caulking (Be75, Wi77). Figure 2.1 shows a typical radon flux sampling measurement using an accumulator can with an attached Lucas Cell. Care must be taken when inserting the accumulator can into the soil so as not to disturb radon in the soil (Pe67). To avoid perturbing the soil-gas, Wilkening (Wi72) proposed inserting a steel ring into the soil initially which can be used for all subsequent measurements. When the radon flux is to be measured, the accumulator can be attached and sealed to the ring without disturbing the soil surface.

When the accumulator is sealed to the surface, the radon concentration entrained within the accumulator can begin increasing. After an adequate accumulation time period, the radon concentration is measured by withdrawing a sample of gas from the accumulator into a measuring cell through a sampling port. A practical method of sampling and measuring the radon concentration is with an evacuated Lucas Cell (Ge76, Lu57, Lu77, Ni81) attached via a sampling port to the accumulator. After about a 4-hour delay, the radioactive daughters will be in equilibrium with the radon. The alpha-decay events within the Lucas Cell are then counted in a light-tight chamber with an alpha scintillation detector. The radon flux is determined from an equation of the form

$$J = \frac{CV}{AT} \tag{1}$$

where

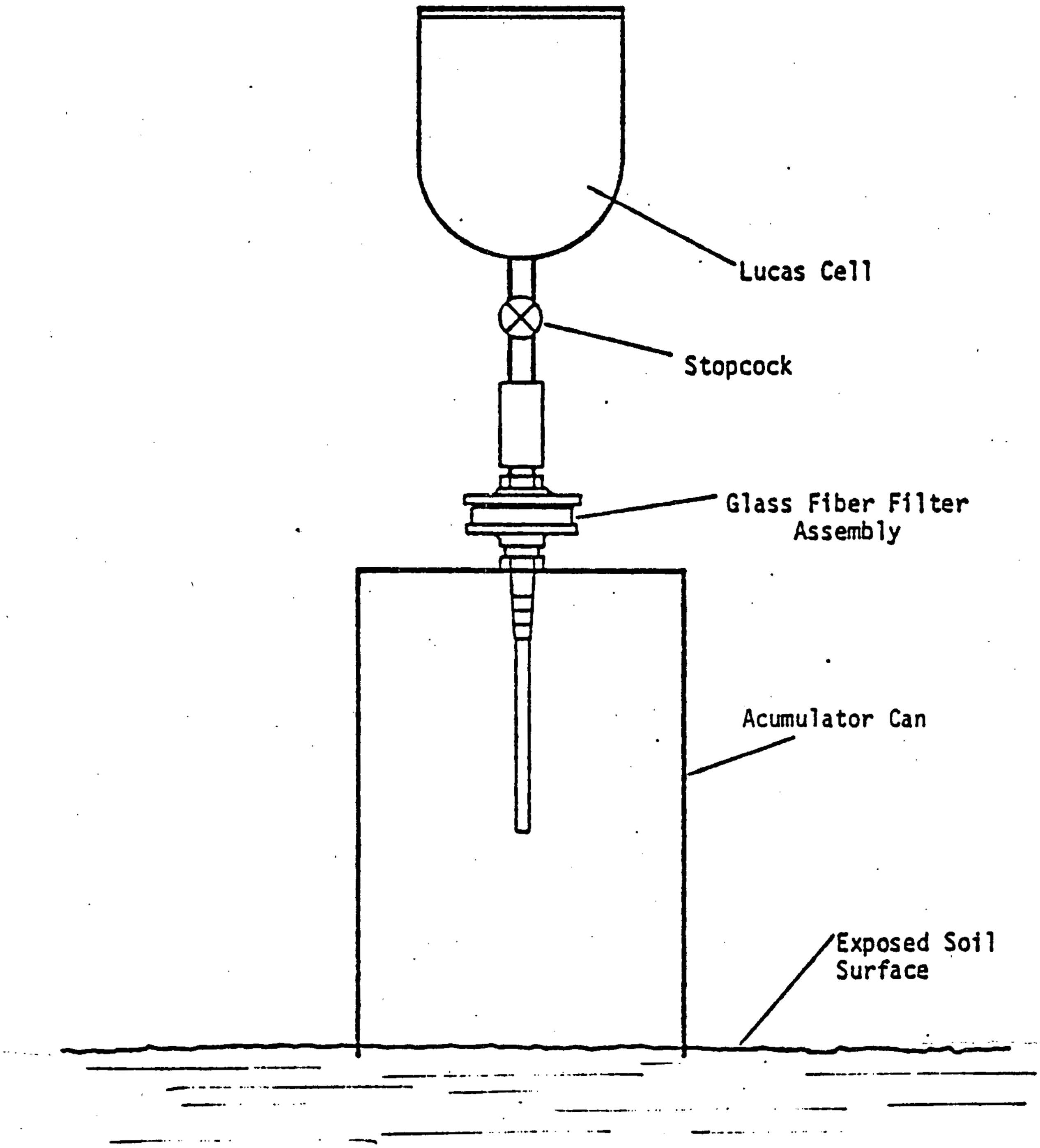


FIGURE 2.1. RADON FLUX SAMPLING WITH ACCUMULATOR CAN AND LUCAS CELL.

$$C = \frac{\lambda c}{eff \cdot v \cdot f\left\{\exp(-\lambda t) - \exp\left[-\lambda(t+\Delta t)\right]\right\}} \qquad (2)$$

and where

J = radon flux (pCi $m^{-2}s^{-1}$)

C = radon concentration in accumulator (pCi/cm^3)

V = volume of accumulator (cm^3)

A = soil surface enclosed by accumulator can (m^2)

T = time accumulator is sealed to the surface before sampling the radon concentration (s)

c = total alpha counts detected

λ = decay constant of radon (2.1×10^{-6} s^{-1})

v = volume of Lucas Cell (cm^3)

t = time between sampling concentration and beginning alpha scintillation counting (s)

Δt = time period of alpha scintillation counting (s)

f = conversion factor: 0.111 alphas/sec per pCi radon

eff = counting efficiency for the Lucas cell

The accumulator should not be sealed to the soil surface for an extended time period. If the radon concentration in the accumulator can becomes too high, then the natural radon concentration gradient in the soil is modified so that some of the radon in the soil beneath the accumulator will move radially through the soil around the accumulator resulting in a reduced concentration measurement. The phenomenon of perturbed radial diffusion has been demonstrated both experimentally and theoretically by Bernhardt, et al., (Be75), Nielson, et al., (Ni81), and Sandquist, et al., (Sa81a). Figure 2.2 provides a comparison of the analytical and experimental measurements which demonstrate the radial perturbation of the radon flux which results from disturbance in the surface radon concentration. The accumulators used in Nielson's experiments demonstrated a 5-percent error in measured concentration when the accumulator was sealed to the soil surface for 10 minutes. The error increased to 11 percent for a 20-minute interval and 33 percent for a 60-minute interval. Using data presented by Nielson based upon diffusion theory, it can be shown that the radon concentration within the accumulator after a 20-minute interval was approximately 40 percent greater than the original soil-gas concentration at the depth to which the can was depressed (1 cm). Figure 2.3 indicates the temporal reduction in radon accumulated in a constant volume sampler with time. Thus, a practical limiting time for accumulation between sampling can be defined

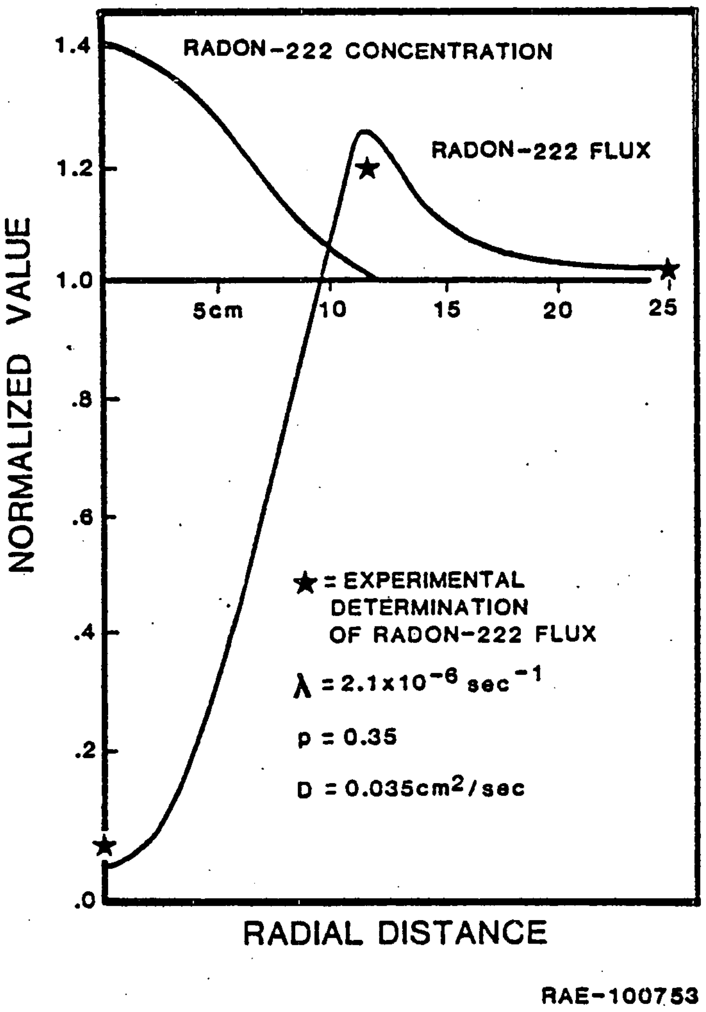


FIGURE 2.2. COMPARISON OF ANALYTICAL AND EXPERIMENTAL NON-UNIFORM RADON-222 FLUX DISTRIBUTIONS.

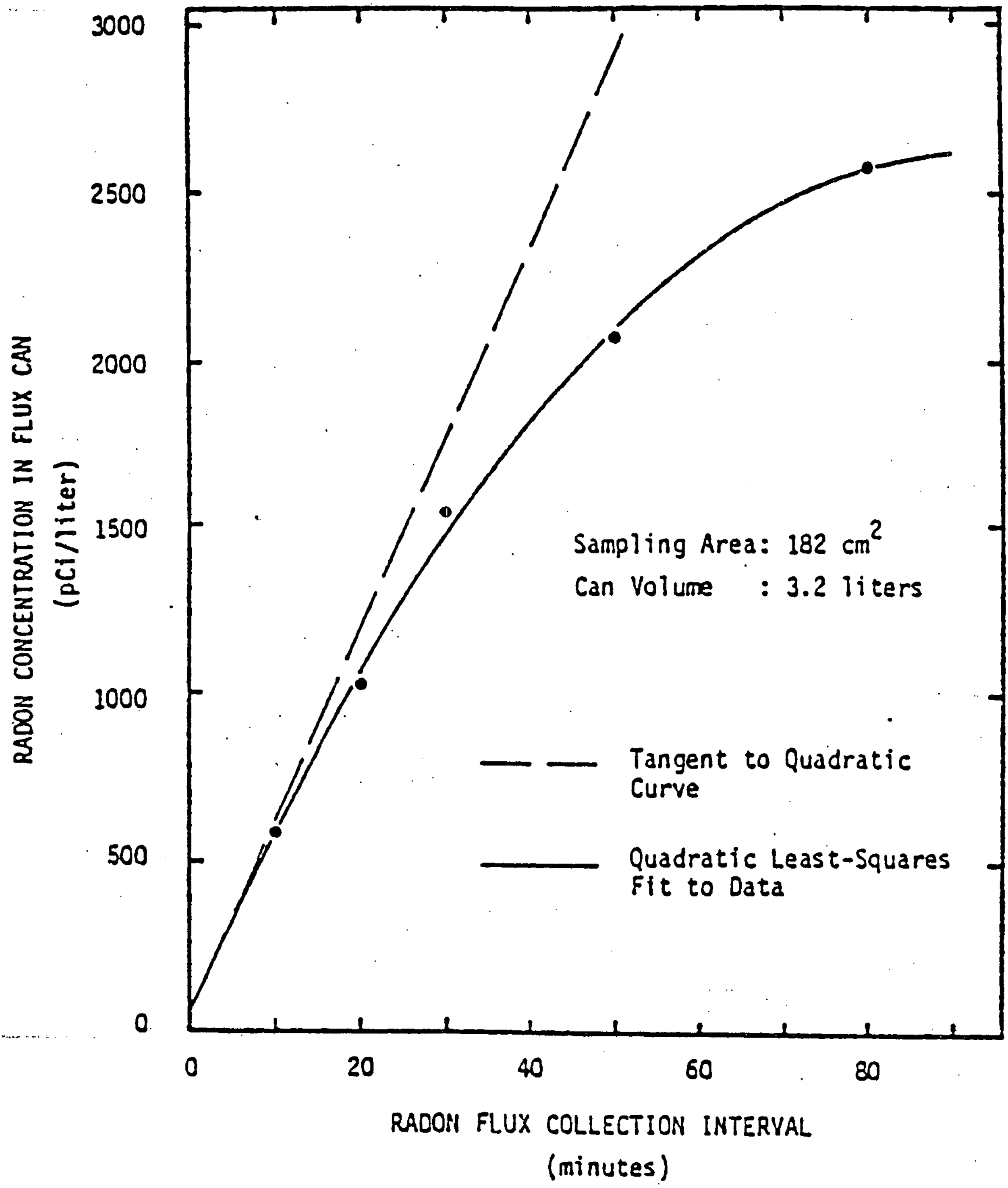


FIGURE 2.3. RADON ACCUMULATION DURING ACCUMULATOR SAMPLING.

as that time when the concentration within the accumulator is equivalent to the original soil-gas concentration at the depth to which the accumulator or ring is depressed. This limiting time can be expressed as:

$$t_L = \frac{xV}{AD_e} \tag{3}$$

where

t_L = sampling time limit (s)

x = soil depth to which accumulator is depressed (cm)

V = volume of accumulator can (cm^3)

A = surface area of accumulator opening (cm^2)

D_e = effective bulk diffusion coefficient of the surface soil (cm^2/sec)

From this equation it should be noted that the limiting time is independent of the magnitude of the flux. The limiting time can be estimated for specific sampling conditions. The dry top surface of a typical soil may have an effective bulk diffusion coefficient in the range of 0.008 to 0.03 cm^2/sec (Ni81, Ro82a, Ro82b). If the accumulator can has vertical sides, $V/A = h-x$, where h is the height, then the limiting time in seconds (for $D_e = 0.02 \ cm^2 \ s^{-1}$) would be:

$$t_L = 50 \ x(h-x) \ \text{(seconds)} \ ,$$

where x and h are measured in cm. A vertical-wall accumulator can 20-cm high inserted to 1-cm depth in the soil would have a limiting time of 950 seconds or about 16 minutes. If the same accumulator were depressed to 2-cm, the limiting time would be 1800 seconds or about 30 minutes. The accumulator can used by Nielson had a height of approximately 17.5-cm and was inserted 1 cm into the soil, which would result in a limiting time of only 13.7 min. As noted (Ni81), 10-min samples were subject to a 5-percent negative bias, which increased to 11-percent for 20-min samples. Thus, this approximate sampling-time criterion helps estimate the time limit beyond which about 7 percent error is introduced into the measurement of radon flux by radial diffusion.

The major disadvantage of the accumulator-can procedure for radon flux monitoring is that the result is valid only for the specific time interval when the measurement is performed. Haghi has indicated (Hag80) that numerous measurements must be taken with this method to determine a reliable average radon flux for a typical disposal site. However, an advantage of this procedure is the relative ease with which accurate measurements can be made. If the accumulator system used is properly calibrated, reliable, instantaneous, radon flux measurements can be obtained. This procedure is particularly useful for determining rapid variations in the radon flux caused by atmospheric or other environmental conditions.

2.3.2 Vertical air column

Wilkening (Wi72) and Bates (Bat81) indicate that the vertical atmospheric profile method of determining radon flux was first suggested by Joly and Smith and has been widely used in the Soviet Union by Malakhov and Kirichenkov. It is assumed that the radon entering a large, vertical column of quiescent atmosphere can be determined from the vertical concentration of radon within that column of atmosphere. However, the method assumes steady-state conditions.

The reliability of the results from atmospheric concentration measurements are significantly affected by air turbulence or vertical or horizontal air currents. Although this method may have some use in determining the average flux from large areas over long time periods, it is easily subject to measurement errors and is not a practical method for determining the flux. In general, other experimental methods of flux measurement are much simpler to perform, and are more reliable and accurate. In addition, this method does not allow for radial diffusion in the atmosphere from small sections of high radon flux to those of low flux; an average flux for the entire region is measured. The size of that region will depend on the elevation above the ground surface at which the measurements are taken.

2.4 INTEGRATED RADON FLUX MEASUREMENTS WITH ACTIVATED CHARCOAL

An integrating radon flux measuring device is one which collects the radon over a moderately long period of time, with the total radon collected being an integrated measure of the temporal flux entering the device. The result of the measurement is an average radon flux for the collection period. The major advantage of integrated measurements over instantaneous measurements is the ability to easily determine an average flux with a single measurement instead of many instantaneous measurements.

The objective of all integrated measurements is to collect radon releases over a period as long as reasonably possible to obtain a truly representative average radon flux for the measurement location. The integrated radon flux measurement method (Al81) most often employed is radon absorption in activated charcoal (Boy07, De03) followed by counting of the entrained radioactive contents (Cou76, Fre81, Me72, Ni81, Ro82b). Nuclear "Track Etch" methods and TLD detectors have also been suggested (Fl80, Fra82) but have not been widely used since such devices measure radon concentration and the radon flux must be inferred from the concentration data.

Activated charcoal has been utilized in various sampling devices and configurations to measure radon flux (Ni81, Cou76, Cou77, Me78, Fre81). The objective is to bring the radon into intimate contact with the activated charcoal. As radon passes over the charcoal the radon is adsorbed onto the charcoal surface. Although most airborne radon is adsorbed onto the charcoal, a small fraction of the radon will remain in the air as defined by the distribution coefficient for the given charcoal type and temperature (Ro82b). It is therefore necessary to avoid exposing the charcoal to the open atmosphere. If the charcoal remains open to the atmosphere the high rate of air exchange around the charcoal will significantly decrease the amount of radon collected by the charcoal, resulting in an erroneous measurement of the flux.

2.4.1 Small-area canister

A simple configuration for radon flux sampling with activated charcoal uses a small gas-mask canister containing activated charcoal (Ni81, Ni83, Thom82), as shown in Figure 2.4. This measurement method is simple, inexpensive and reasonably accurate. The small charcoal canister can be easily covered and sealed with an empty, 8.2-cm by 4.1-cm, open-top can (e.g., a standard, half-pound, tuna fish can is about this size). If the can is gently inserted into the surface soil to a depth of 1 cm, then the maximum sampling time is approximately 31 hours (see Section 2.6.2 on sampling times). An integrated flux measurement can, therefore, readily be taken over a one-day period, resulting in a representative measurement of the average daily radon flux at that position. After the radon sample is collected in the charcoal canister, the canister is enclosed and sealed in an airtight, impermeable container such as a sealable plastic bag or metal can for approximately four hours while the radon and its decay products come into radioactive equilibrium. The canister is then counted for activity in a calibrated gamma radiation detector (NaI or Ge(Li) which is set to count the 609-keV gamma-ray photopeak produced by the decay of bismuth-214. The radon flux can be calculated using the following equation:

$$J = \frac{c\lambda^2}{\mathrm{eff} \cdot A \cdot \left[f\ 1-\exp(-\lambda t_2)\right]\left\{\exp\left[-\lambda(t_3-t_2)\right]-\exp\left[-\lambda(t_4-t_2)\right]\right\}} \qquad (4)$$

where

J = radon flux (pCi $m^{-2}s^{-1}$)

c = net observed counts for Bi-214 photo peak

λ = radon decay constant (s^{-1})

t_2 = radon flux sample collection interval (s)

t_3 = time from start of sample collection to start of count (s)

t_4 = time from start of sample collection to end of counting interval (s)

A = area of canister exposed to radon flux (m^2)

eff = detector efficiency (counts/disintegration)

f = unit conversion factor: 0.037 picocuries per disintegration-per-second

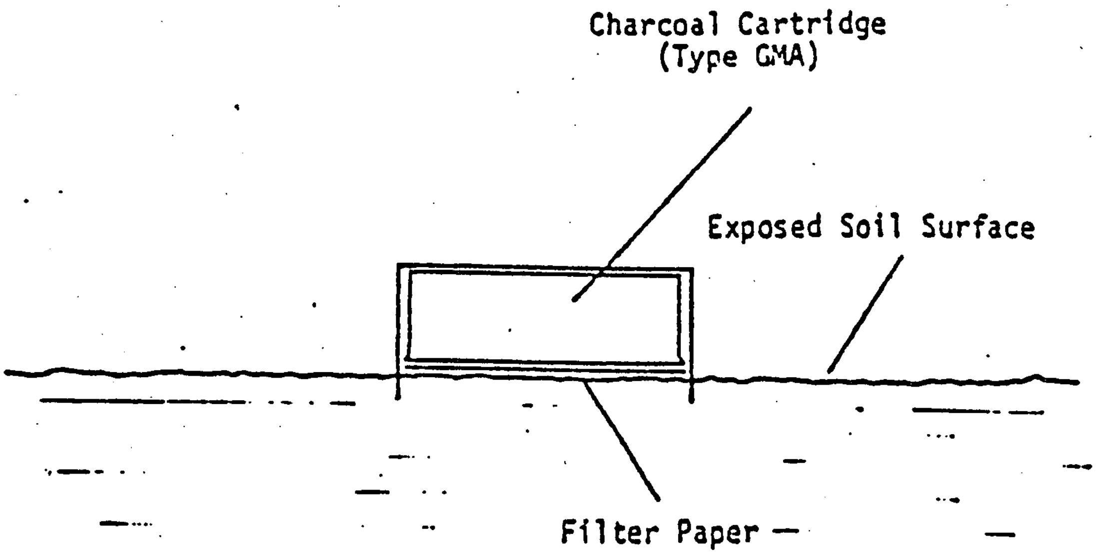


FIGURE 2.4. RADON FLUX SAMPLING WITH A CHARCOAL CANISTER.

The major advantages of the small charcoal canister are the simplicity of field sampling and the relatively long integrating time for sampling. The canisters can be readily placed and collected routinely by nontechnical personnel and transported for gamma-counting. The major disadvantage is the small sampling area of the canister, which can lead to imprecision due to spatial variability in flux resulting from buried rocks or other local anomalies; however, as discussed in Section 2.6.1, this disadvantage is easily compensated for.

2.4.2 Large-area passive canister

A large-area, passive sampler has been designed (Ri81) to achieve the benefits of large-area sampling and yet reduce the complexity of the charcoal tent air-flow system discussed in Section 2.4.3. A large-area, passive sampler can be easily prepared by placing activated charcoal in a permeable bag which can be spread over the surface area to be sampled. The surface of the bag in contact with the soil must be porous to radon while the upper atmospheric surface of the bag must be covered with an impermeable cover which is pressed gently into the soil and sealed or connected to an appropriate size ground ring as previously discussed.

The maximum radon collection time can again be determined from the equilibrium ratio. After the appropriate collection interval, the bag of activated charcoal is then carefully placed in an impermeable container and sealed. After a minimum of four hours, the radon is analyzed as described in Section 2.4.1.

The physical arrangement of the charcoal spread over the surface under an impermeable cover can, of course, be varied as desired. The objective of this counting system is to spread the charcoal over as large an area as is practical, and sample for the radon flux for the maximum time practical. The major advantage of the passive canister system is the large area which can be sampled for radon flux without the complexity of mechanical equipment. A major disadvantage is the additional care and time required in adequate setup of the sampler in the field.

2.4.3 Charcoal tent

A larger charcoal sampler for monitoring radon flux has been designed (Ni79, Fre81, Thom82), and is referred to as a charcoal tent. This sampler improves the radon spatial averaging capability, and thus reduces the possible effects of soil inhomogeneities beneath the measurement system (Fre81). As presently implemented, the tent is a large, flat metal cover which contains an elaborate system for air circulation and pressure equilization within the cover (Har81) (see Figure 2.5 for details). Soil gas containing radon passes under the tent and through charcoal filters where the radon is adsorbed. The length of the effective radon collection period is controlled by the amount and mesh size of the activated charcoal in the filter and the ratio of charcoal to air in the system. Utilizing the distribution coefficient between charcoal and air as defined in Section 2.4.1, the effective collection period can be defined, and is generally a period of a few hours.

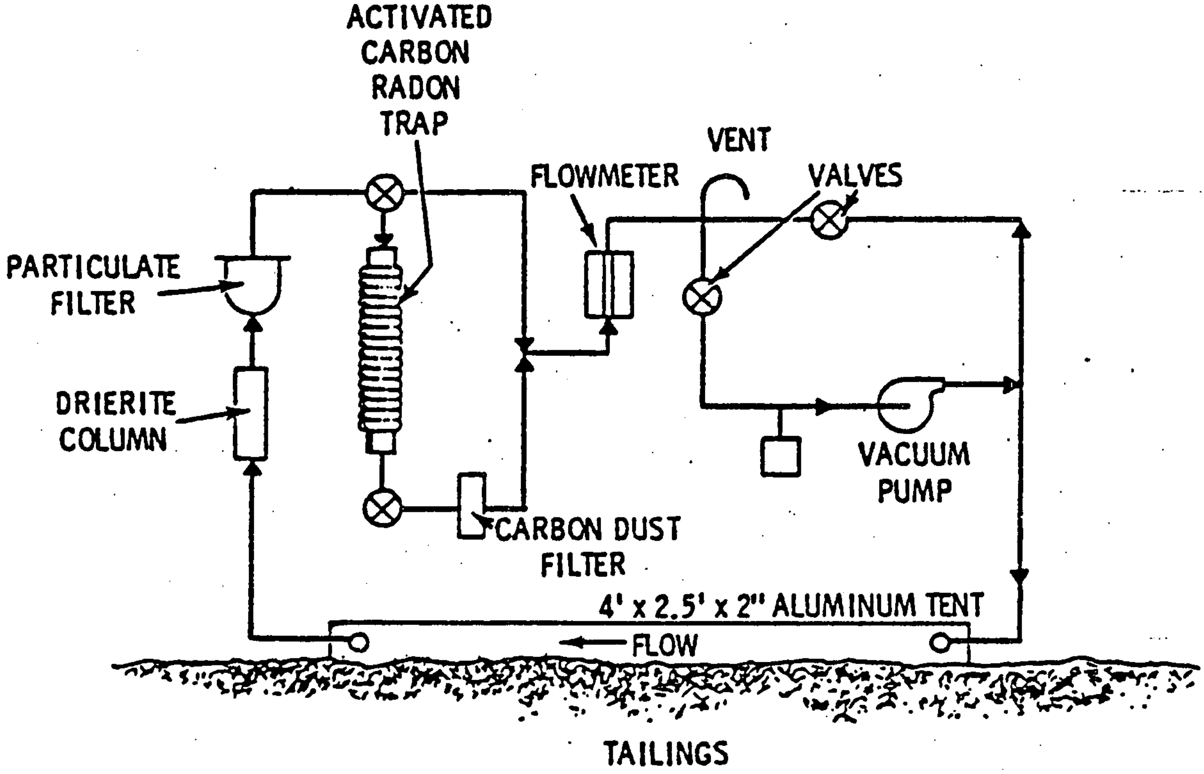


FIGURE 2.5. RADON FLUX FIELD MEASUREMENT SYSTEM. (Fre81)

After an appropriate collection period, the charcoal is placed in a sealed container. After a delay of at least four hours for radiological equilibration, the radon on the charcoal is counted in the same way as for the small canister described in Section 2.4.1.

The major advantage of the charcoal tent over the gas-mask canister is the larger area sampled. The major disadvantage is the complexity and cost of utilizing the system.

2.5 CONTINUOUS RADON MONITORING

Continuous monitoring of the radon flux through soils has been successfully performed with alpha detectors using a flow-through, radon-flask detection system in which air is continuously passed from a sampling chamber to the detector (Au81, Mom79b). Continuous radon concentration monitors that could potentially be used in similar continuous flux monitoring instruments have also been reported, utilizing both radon flasks (Cou77, Ge76, Tho79) and a two-filter system (Ha78, Tho70). The major advantage of any continuous monitoring system is its ability to measure the radon flux through all time variations experienced at a location. The average radon flux over any given time period can then be determined by averaging the results for the appropriate sampling-time interval. The major disadvantage of continuous monitoring systems is the complexity of the sampling operation and the equipment required. Although the occasional need for continuous radon measurements may warrant the use of a continuous radon flux monitoring system, the use of such equipment and its operational demands should be weighed against simpler and less costly methods available from integrated and grab sampling methods.

2.6 ERRORS AND UNCERTAINTIES IN EXPERIMENTAL METHODS

This section reviews the sources of errors, uncertainties and sensitivities of the various methods and procedures for measuring radon flux to help define their capabilities and limitations. These parameters are important in establishing procedures for measuring radon flux which will satisfy the objectives of the measurement program.

2.6.1 Temporal and spatial measurement requirements

The determination of accurate radon flux measurements at a specified site can be markedly affected by both time- and space-dependent factors, as described in Section 2.2. Thus, to make an accurate assessment of the actual radon flux releases, it is important to design an adequate flux monitoring procedure, as well as to select a satisfactory measuring device.

In a statistical analysis of temporal uncertainties from radon flux measurements, Haghi, et al., (Hag80) state that it may be necessary to perform over one hundred independent, instantaneous measurements of the radon flux to restrict the maximum measurement error to 5% (with a 95% confidence level) for the actual time-averaged flux. Haghi further indicates that under some highly time-varying conditions, a maximum 5 percent error in the actual flux may require over one thousand instantaneous measurements. It is evident, in view of the large number of measurements required for accurate determination of the time-average radon flux by instantaneous sampling devices, that integrated and continuous flux measuring systems enjoy a significant advantage. Even for these measurement methods, it is important to remember that the radon flux can vary not only diurnally but also seasonally. The ideal time-averaging period for temporal flux measurements is a calender year which has exhibited typical climatic conditions (precipitation, temperatures and pressures).

To provide reasonable spatial averaging measurements of the radon flux from a given site, Leggett et al., (Le78) have suggested that spatially varying experimental parameters such as the radon flux should be measured at the following number of independent (random or uniformly distributed) locations:

$$\text{Number of locations} = 45\ (\text{coefficient of variation})^2 \qquad (5)$$

to achieve a precision of 25 percent or less uncertainty (for a 95 percent confidence level). However, he recommends that a minimum of 30 measurements be made even if the coefficient of variation suggests a smaller number. Figure 2.6 provides a simple graph of the number of independent spatial measurements required. For example, the radon flux measurements performed by Ford Bacon & Davis Utah, Inc. (FBDU), for several uncovered mill tailings sites provided an average coefficient of variation of 0.84, which implies that a minimum of 32 independent spatial measurements should be made to establish the average spatial flux to within 25 percent uncertainty of the actual spatial average value.

In a recent analysis of precisions in radon flux estimates (You83), the effect of the sampling area was examined. Using a large (0.929 m^2) tent sampler, the spatial coefficient of variation for the Grand Junction tailings pile was indicated to be 0.84, suggesting the need for 32 samples (Equation 5) to define the average flux. From the data using smaller (41 cm^2) samples, a coefficient of variation of 0.29 was estimated for an area the size of the large tent sampler. The overall coefficient of variation for the pile, if the small sampler had been used, was then estimated to be $[(.84)^2 + (.29)^2]^{½} = 0.89$, which corresponds to a requirement of 36 samples (Equation 5). If this analysis is correct, there is little to be gained in using large-area samplers. Data from various flux samplers on a less homogeneous source suggest that up to three times more small samplers may be needed (Thom82) to give precisions equivalent to those from larger samplers, however, so the equivalent numbers of small samplers are not presently well defined.

2.6.2 Activated charcoal

The determination of the radon flux utilizing charcoal adsorption is based upon experimental measurements which can be made both precisely and accurately. As illustrated in Equation 4, the quantities measured are the soil surface area being sampled, the gamma photopeak counts measured for Bi-214, the sampling, decay and counting time intervals, and the efficiency of the detector system.

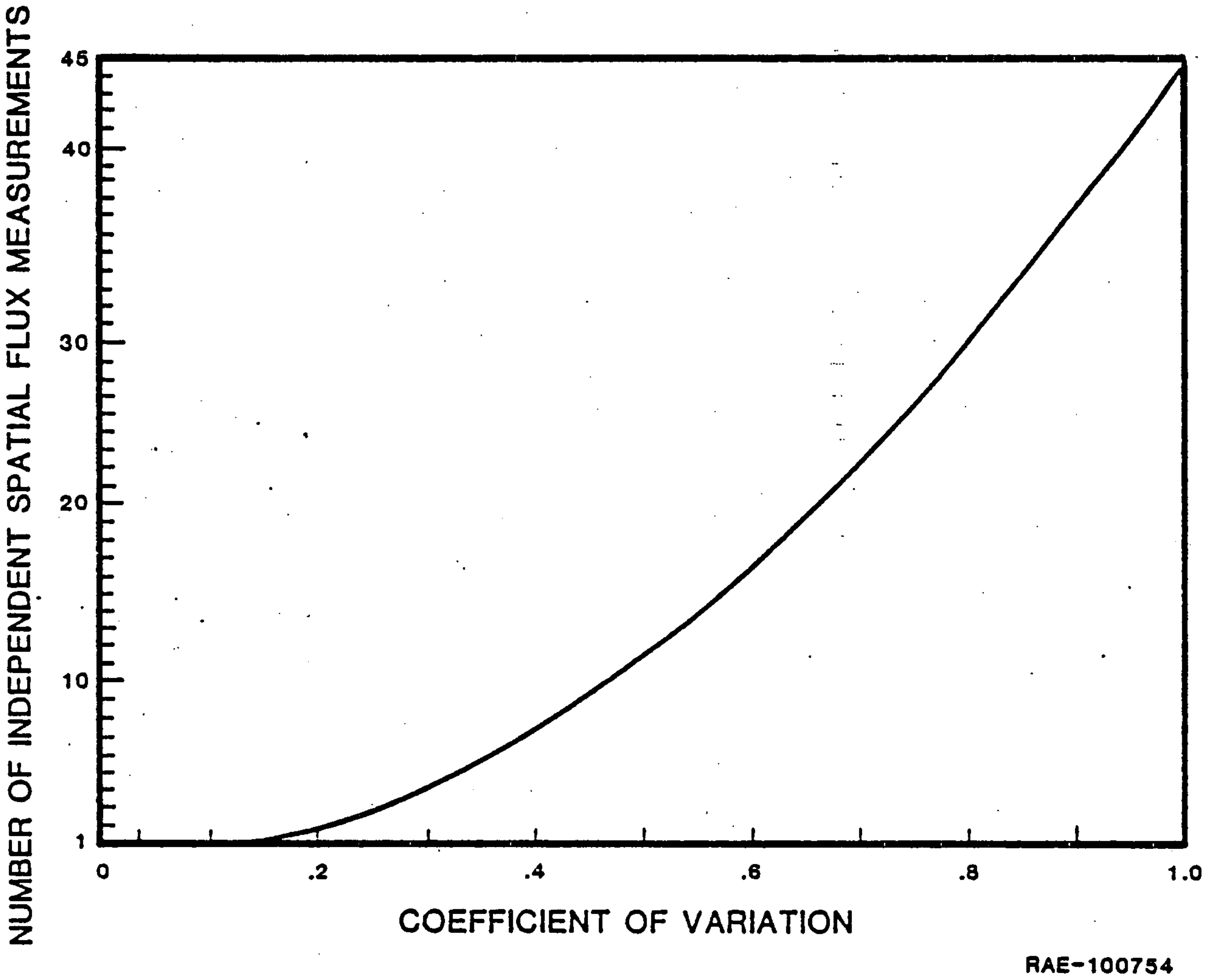


FIGURE 2.6. THE NUMBER OF INDEPENDENT SPATIAL MEASUREMENTS OF THE RADON FLUX VERSUS THE COEFFICIENT OF VARIATION THAT ARE REQUIRED FOR A 25% MAXIMUM ERROR.

The uncertainty in the times, as well as in the area of the cover, can be made minimal. The experimental uncertainty in the radon flux measurement, therefore, can be expressed approximately as:

$$\frac{\Delta J}{J} = \left[\left(\frac{\Delta eff}{eff}\right)^2 + \left(\frac{\Delta c}{c}\right)^2 \right]^{\frac{1}{2}} \qquad (6)$$

If the detector efficiency is accurately measured, the relative error in the radon flux measurement approaches the relative error in the total gamma counts accumulated. This counting error can be reduced if necessary by extending the counting time. Thus, the uncertainty in the measured flux can generally be controlled by accurate determination of the counting system efficiency, sufficiently long counting times, and care in making the measurements within acceptable procedure limitations.

One limitation which can often become a dominant source of error is the decline in adsorption efficiency after the charcoal has already adsorbed large quantities of radon. The charcoal does not adsorb all radon entering the sampler; a small portion remains in interstitial air surrounding the charcoal. The ratio of the adsorbed radon (pCi/g) to the gaseous radon concentration is defined as the distribution coefficient of the charcoal, from which the equilibrium gaseous radon concentration can be estimated. After long sampling times, the gaseous radon concentration can become large, causing radon in the soil to begin to diffuse radially around the sampler to the soil surface where there is a stronger gradient for radon diffusion. The result is a reduced radon flux measured in the collection system, as demonstrated by Figure 2.7 for bare and covered tailings systems.

To determine the time limitation of the monitoring system, it is necessary to define the equilibrium radon distribution coefficient between the activated charcoal and the air. This coefficient, K, has been experimentally determined at RAE for activated carbon (Marine Enterprises, Towson, MD), as shown in Figure 2.8 and mathematically defined as follows:

$$K = 9620 \times 10^{-0.017T} \left(\frac{pCi/g}{pCi/ml}\right) \qquad (7)$$

where T is the ambient air temperature in ^{o}C.

The total radon activity entering the collector system, the sum of the activity in the air and the activity on the charcoal, can be expressed as:

$$R_T = JAt = R_C + R_A \qquad (8)$$

where

R_T = total radon activity (pCi)

J = radon flux (pCi $m^{-2}s^{-1}$)

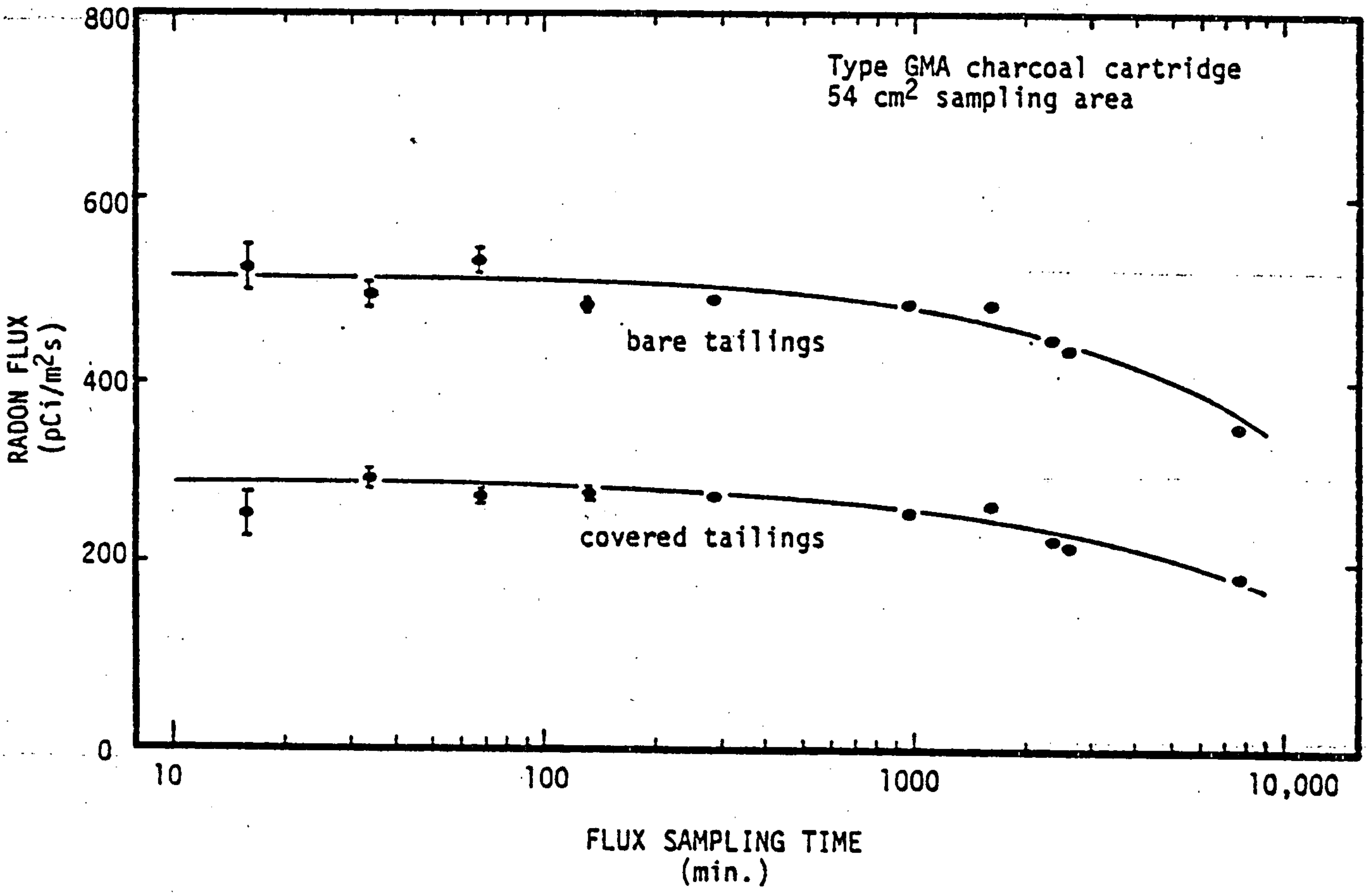


FIGURE 2.7. VARATION IN RADON FLUX SAMPLING EFFICIENCY WITH SAMPLING TIME.

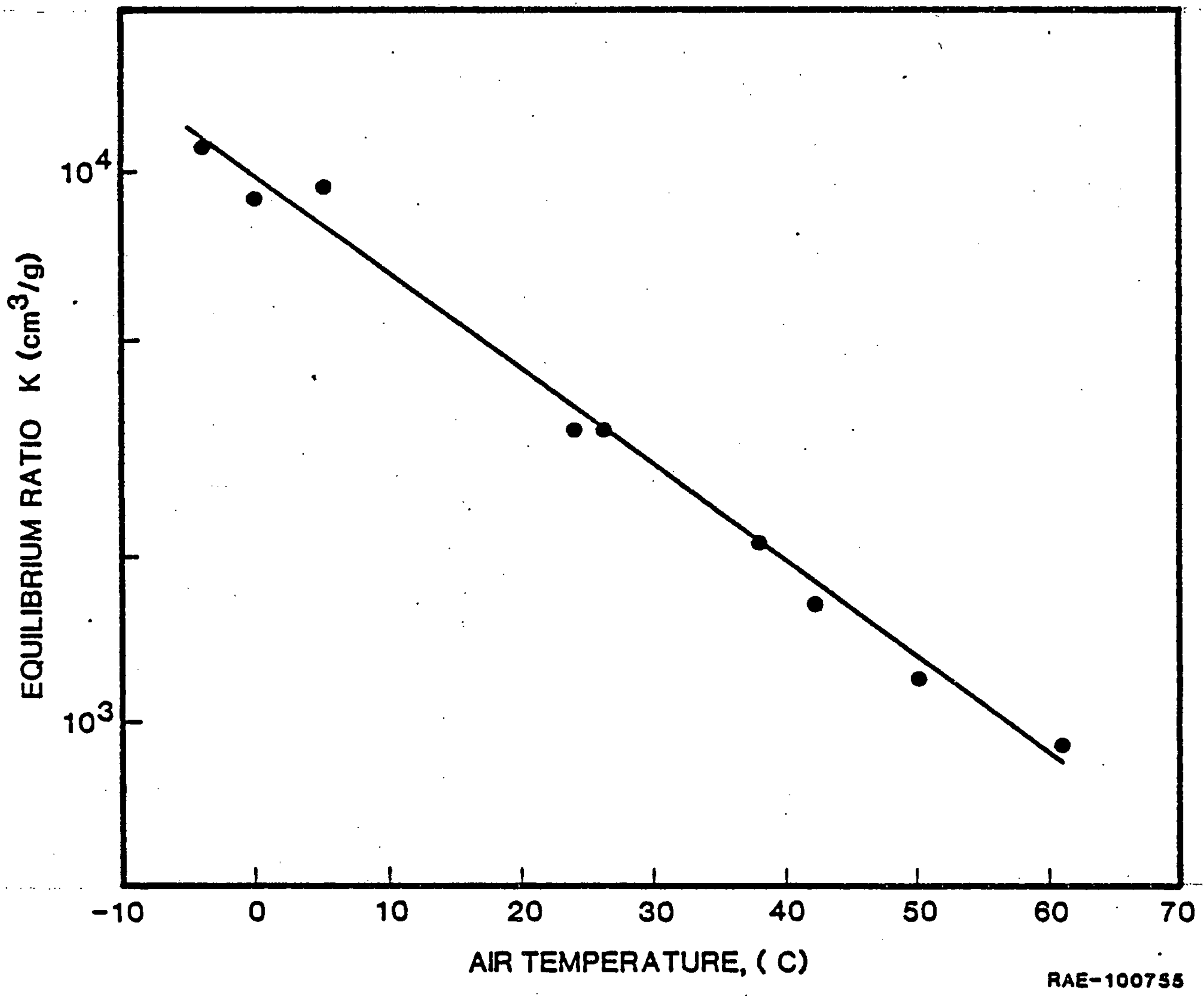


FIGURE 2.8. THE RADON EQUILIBRIUM RATIO AS A FUNCTION OF AMBIENT AIR TEMPERATURE.

A = open area of collection system (m^2)

t = time period of collection (s)

R_C = radon activity on the charcoal (pCi)

R_A = radon activity in the air (pCi)

Substituting for the radon activity on the charcoal and in the air, the total radon activity can be expressed as:

$$JAt = C_a (km + V_a) \tag{9}$$

where

C_a = radon concentration in the air (pCi/cm^3)

k = radon distribution coefficient for charcoal/air (cm^3/g)

m = mass of charcoal (g)

V_a = air volume in the system (cm^3)

The time limit for collection can therefore be expressed as:

$$t_L = \frac{C_{am}}{JA} (km + V_a) \tag{10}$$

where

t_L = maximum efficient collection time (s)

C_{am} = maximum air concentration (pCi/cm^3)

If the maximum air concentration is taken to be the approximate soil-gas concentration at the depth to which the collection system is depressed (see Section 2.3.1 and Ni81) the maximum air concentration can be expressed as:

$$C_{am} = 10^{-4} \frac{Jx}{D_e} \tag{11}$$

where

x = depth to which collector is inserted (cm)

D_e = effective bulk diffusion coefficient (cm^2s^{-1})

J = radon flux (pCi $m^{-2}s^{-1}$)

10^{-4} = units conversion factor (m^2/cm^2)

Substituting Equation 11 into Equation 10 for the maximum efficient collection time, it can be seen that the maximum time is independent of the value of the radon flux, and is given by:

$$t_L = \frac{10^{-4}x}{AD_e}(km + V_a) \qquad (12)$$

Equation 12 gives the approximate maximum efficient collection time for a charcoal collection system. It does, of course, assume that there is no impediment to maintaining the radon equilibrium ratio between the charcoal and the air at the soil surface at all times. If a significant delay is required for the radon to reach the charcoal, as may occur due to an air gap between the soil and charcoal, then the air concentration will build up faster and reduce this maximum efficient collection time.

In conclusion, the uncertainty in flux measurements with activated-charcoal-based measurement systems can be expressed by Equation 6, if the sample-collection period is maintained less than the maximum efficient time, as expressed by Equation 12. Utilizing reasonable care with instruments properly calibrated, the radon flux should be measurable within a few percent using a charcoal collection system. Excessive moisture accumulation on the charcoal will lower its radon distribution coefficient, and hence the value for t_L (Ni81). Although the radon flux samplers are not highly sensitive to advective movement as induced by barometric pressure changes (Ro83), pressure variations strongly affect the instantaneous flux, and usually require longer sampling periods to average over short-term anomalies.

2.6.3 Alpha scintillation

The sensitivity of the scintillation method of measuring radon can be contolled within acceptable limits by utilizing proper procedures and calibrations. Thomas and Countess (Tho79) indicate that the relative error of the continuous radon monitor used in their laboratory can be expressed as:

$$E_r = \frac{1.09}{C}(175C + 1.5B)^{1/2} \qquad (13)$$

where

E_r = relative error

C = radon concentration in pCi/l

B = background counts during the counting interval

However, Williams, et al., (Wil81) indicate that there are empirical errors as well as theoretical errors which must be considered. In reviewing measurements made in international laboratories, their conclusion was that a precision of 7 to 12 percent uncertainty was possible with either 200 counting events and/or a counting time of 200 minutes.

2.7 CONCLUSIONS

Both instantaneous and integrated measurements of the radon flux have been successfully made using the accumulator-Lucas cell and activated charcoal methods. The instantaneous grab samples are effective in following flux variations, while the measurement methods based upon charcoal adsorption of radon provide a better daily average. The Lucas-cell grab sample is limited to a short accumulation time, as defined by Equation 3. The charcoal integrated sample can average the measurement over approximately 24 hours, with the maximum time (not including the limitation caused by radon decay) defined by Equation 12. Within the appropriate collection-time limitations, both of these methods should provide acceptable measurement precision of the radon flux.

3.0 THEORETICAL AND COMPUTATIONAL MODELS FOR RADON FLUX DETERMINATIONS

In this chapter the theoretical and computational models for determining radon flux are reviewed with regard to applications to uranium mill tailings sites.

3.1 BACKGROUND

The various physical processes by which radon can be transported from one point to another are numerous, interactive and complex. In general, however, these processes may be categorized for mathematical modeling as macroscopic transport processes and microscopic transport processes (Har38, Ni82b, Ro81b, Ro83, Sc82a, Ta64, Ta80, Wi60, Yu81).

Macroscopic transport accounts for those transport processes in which radon atoms become intimately associated with their surrounding medium which itself is undergoing transport through the medium. For example, the entrapment of radon in soil, water or air and the subsequent flow of that water or air through the soil is a macroscopic radon transport process. In the overall movement of radon in containment systems, both macroscopic and microscopic processes can be important, but they are usually identified and evaluated separately. Evaluations have shown that the macroscopic transport from diurnal atmospheric pressure variations is negligible when averaged over long periods (Bav56, Ro83).

Microscopic transport of radon is that collection of processes by which individual atoms of radon move as a consequence of momentum, thermal or mass gradients imposed upon the spatial radon distribution. Molecular diffusion of radon is one type of microscopic transport process. It results when radon atoms exhibit a variation in concentration, causing a net flow of radon to reduce the concentration gradient. This simple molecular diffusion is generally modeled mathematically by Fick's law.

If a temperature or momentum gradient exists within a containment volume which includes radon, then a microscopic transport of radon will arise to minimize these gradients also. However, microscopic transport of radon arising from thermal or momentum gradients is generally cyclic and assumes an average value which is small over long time periods in uranium tailings cover systems. Therefore, thermal and momentum gradients may usually be ignored.

3.2 MATHEMATICAL MODELS FOR RADON TRANSPORT

A large number of theoretical and analytical studies (Bat80b, Du76, Ed80, Go80, Je80, Ka78, La82, Ro82a) have been devoted to modeling and analyzing the transport of radon through various media and geometries under a variety of assumptions (e.g., nonhomogeneous media, time-dependence, moisture effects, advective flow, soil surface fissures, and nonuniform radon source distributions.) and boundary conditions. The results and findings of these studies are primarily useful in designing more accurate measurement experiments, explaining experimental anomolies, and providing an improved understanding of the radon transport problem. The following equations give several of the general mathematical descriptions and the standard solutions recognized and employed in simple radon transport analysis.

The space- and time-dependent transport of radon through a material may be mathematically modeled by the radon balance equation (Bav56, Is70, Me73, Ro83, Tu77) which sets the total time rate of change of the radon concentration C(r,t) equal to the sum of the sources and losses of radon as follows:

$$\frac{dC(\bar{r},t)}{dt} = \frac{\partial C}{\partial t} + \nabla \cdot \frac{\bar{J}}{p} = Q - \lambda C \tag{14}$$

where

$C(\bar{r},t)$ = the radon concentration in the porous space at vector position $\bar{r}$ at time t (pCi/cm^3)

∇ = the spatial divergence operator (cm^{-1})

$J(\bar{r},t)$ = the vector current (flux) of radon at $\bar{r}$ and t (pCi cm^{-2}s^{-1})

$Q(\bar{r},t)$ = the volumetric radon source strength in the porous space at $\bar{r}$ and t (pCi cm^{-3}s^{-1})

λ = the radon decay constant (sec^{-1})

p = the porosity of the porous transport media (cm^3/cm^3)

The mathematical model for the description of the radon vector current accounting for both macroscopic and microscopic transport (Jo60) is as follows:

$$\frac{\bar{J}}{p}(\bar{r},t) = -D\nabla C + \bar{v}C \tag{15}$$

where

D = the radon diffusion coefficient in the porous space (cm^2/sec)

$\bar{v}$ = the effective macroscopic radon transport velocity in the porous space (cm/sec)

∇ = the spatial gradient operator (cm^{-1})

Equation 15 is modeled assuming that all microscopic and macroscopic radon transport processes can be adequately described by the two terms on the right side of Equation 15. This assumption implies that the parameters D and v may be space- and time-dependent.

Equations 14 and 15 may be combined to eliminate the vector radon current J and provide the following radon transport equation:

$$\frac{\partial C}{\partial t} = \nabla \cdot D\nabla C - \nabla \cdot \bar{v}C - \lambda C + Q \tag{16}$$

Equation 16 with space- and time-dependent parameters is not generally amenable to analytical solution. If approximate solutions of Equation 16 are required, the most practical approach is numerical computer solution of the equation for the specific radon transport system under investigation.

Various analytical solutions (Ro81b, Ro82a, Ro83) for reduced and simplified forms of Equation 16 do exist, however. For example, if the radon transport system under study can be adequately modeled by one-dimensional, steady radon transport with D, v and Q assumed to be constant both in time and space, then Equation 16 reduces to:

$$D\frac{d^2C}{dx^2} - v\frac{dC}{dx} - \lambda C + Q = 0 \tag{17}$$

The advective component in Equation 17 can be neglected for almost all practical applications (Ro83). This yields the traditional one-dimensional, steady-state radon diffusion equation:

$$D\frac{d^2C}{dx^2} - \lambda C + R\rho\lambda E/p = 0 \ , \tag{18}$$

where the source term, Q, is given by $R\rho\lambda E/p$ and

R = specific activity of radium in the soil (pCi g^{-1})

ρ = dry bulk density of the soil (g cm^{-3})

E = radon emanation coefficient (dimensionless)

The radon flux from the bulk soil material, given in Equation 14 reduces to:

$$J = -10^4\, Dp\frac{dC}{dx} \ , \tag{19}$$

where

J = bulk radon flux ($\text{pCi m}^{-2}\ \text{s}^{-1}$)

10^4 = factor to convert units from $\text{pCi cm}^{-2}\text{s}^{-1}$ to $\text{pCi m}^{-2}\text{s}^{-1}$

The solution of Equations 18 and 19 for the flux from a bare, homogeneous tailings pile is:

$$J_t = 10^4\, R\rho E\sqrt{\lambda D_t}\,\tanh\left(x_t\sqrt{\lambda/D_t}\right), \tag{20}$$

where

J_t = radon flux from the tailings surface (pCi $m^{-2}s^{-1}$)

x_t = thickness of tailings (cm)

10^4 = factor to convert units from pCi $cm^{-2}s^{-1}$ to pCi $m^{-2}s^{-1}$

The subscript "t" refers to the tailings region.

The exact solution from diffusion theory to the two-region, tailings-cover problem is (Ro84):

$$J_c = \frac{2J_t\, e^{-b_c x_c}}{\left[1 + \sqrt{a_t/a_c}\, \tanh(b_t x_t)\right] + \left[1 - \sqrt{a_t/a_c}\, \tanh(b_t x_t)\right] e^{-2b_c x_c}} \qquad (21)$$

where

b_i = $\sqrt{\lambda/D_i}$ (i = c or t) (cm^{-1})

a_i = $p_i^2\, D_i \left[1 - (1-k)\, m_i\right]^2$ (cm^2s^{-1})

m = $10^{-2}\rho M/p$ = fractional moisture saturation (dimensionless)

M = moisture content (dry weight percent)

k = 0.26 pCi/cm^3 in water per pCi/cm^3 in air

The subscript "c" pertains to the cover region, and J_t is given in Equation 20.

Solutions to the diffusion equation for more than two regions involve complex sets of coupled equations which are amenable to computer solution using matrix techniques (Ro84).

3.3 COMPUTATIONAL MODELS AND COMPUTER CODES

A broad variety of computational models for predicting the transport of radon through various media, geometries, and boundary and initial conditions exist. For many of the models, appropriate computer codes have been developed and used for predicting and evaluating radon transport effects. Only those codes that have been reasonably well documented and verified will be reviewed here.

3.3.1 RAE radon transport codes

Several major algorithms and computer codes have been developed, documented and published by RAE (Ro80, Ro81a, Ro81b, Ra82c, Ro83, Gr83) under the UMTRA Program which predict the radon flux (and concentration) transported through porous media.

RAECO (Ro81a) - The first fully-documented and published computer code under the UMTRA Program for describing the one-dimensional diffusion of radon through porous media was the RAECO Code. This code combined an analytical, exact solution for the steady-state, one-dimensional diffusion of radon through a multilayer tailings and cover system with a cost optimization model for engineering analysis and design.

This FORTRAN-coded program permits any finite number of one-dimensional layers to be specified, and permits specification of the entrance flux at the bottom of the multilayer system and the surface radon concentration at the soil-atmosphere interface.

Later modifications to the code permit the input of unlimited thicknesses of tailings, and by employing double-precision routines, the minimum range for the radon diffusion coefficients has been reduced to about 10^{-8} cm^2/sec. Recent modifications of the code under the title ADVECT allow for advective movement of radon and also permit interactive modeling and analysis of multilayer tailings cover systems.

3DRD (Sa81b) - This FORTRAN computer code determines the steady-state radon flux and concentration within a heteorogeneous, porous diffusion medium with one-, two- or three-dimensional geometry. The code is based upon a variable-spaced, finite-difference model and solves the resulting difference equations using a Gauss-Seidel accelerated iteration technique. The code permits the specification of the radon flux or concentration distribution at any external boundary surface, and the resulting radon flux is determined at each boundary where a radon concentration distribution was specified.

GASFLX - A time-dependent, one-dimensional radon transport computer code which includes molecular diffusion (Fick's law) and advective transport has been coded in FORTRAN under the name GASFLX. The code algorithm is based upon a direct solution of the tridiagonal matrix equation for the model difference equations.

2DTR - The FORTRAN code 2DTR is a two-dimensional, time-dependent radon transport code which models radon diffusion and advection in two dimensions in rectangular geometry and the radial-axial dimensions in radially symmetric cylindrical geometry. The parabolic, partial differential equation for the model is converted to a finite-difference equation in space. The resulting time-differential, space-difference equation is then solved for the system's time behavior via an analytical algorithm, and the remaining coupled, matrix difference equations for space are then solved explicitly. The code has been employed to evaluate and confirm experimental transient measurements of radon diffusion coefficients and other modeling efforts.

3.3.2. PNL radon transport codes

Three different computer codes have been developed at the Pacific Northwest Laboratory which model radon diffusion through multilayer, porous media.

RADON1 (May 81b) - This code is based upon a one-dimensional, steady-state diffusion model for radon transport. Multilayer cover and tailings systems can be studied and standard boundary conditions imposed (viz., either the radon flux or concentration can be specified at the entrance and exit layers). The code is apparently similar to RAECO, but has not been publicly released.

RNDI1D (May81b) - This code solves one-dimensional, time-dependent radon diffusion problems using finite-difference techniques with an implicit time-difference scheme. It has been used along with moisture-balance calculations to simulate varying radon fluxes resulting from changing soil moisture conditions.

RADMD (May81b) - This code is a modification of the three-dimensional, time-dependent, heat-flow code TRUMP developed at Lawrence Livermore Laboratory by A.L. Edwards (Ed72). The RADMD will model steady-state or time-dependent radon diffusion in rectangular, cylindrical, or spherical geometry for one, two, or three dimensions. The computational algorithm is based upon an integrated, finite-difference representation of the governing partial differential equation. The code will accommodate mixed boundary conditions, i.e., a distribution of radon flux or concentration values over a control surface. Although output data from the code have been published, no source listing or public release of the code is available.

3.3.3 ANL radon transport code (Yu81)

A computer code has been developed for the one-dimensional transport of radon in a multilayer porous medium that yields a time-dependent estimate of the surface radon flux. The code is based upon the mathematical model of Clements and Wilkening and uses Darcy's law to account for soil-gas movement due to atmospheric pressure changes. The Galerkin method is employed to provide finite-element discretization for the model. Then the resulting time-dependent matrix equation is solved using the Crank-Nicholson finite-difference method. Boundary conditions employed are zero radon concentration and atmospheric air pressure at the soil surface.

No public releases of the source code are available.

3.3.4 Bureau of Mines radon transport code (Bat78, Bat80a, Bat80b, Bat81)

A mathematical model and computer code have been developed by Bates and others at the Bureau of Mines for estimating radon emanation into underground uranium mines. The computer code can simulate the one-dimensional, time-dependent transport of radon driven by both diffusion and advection arising from variations in mine ventilation pressure. The advective velocity is determined from a Darcy flow model.

The radon transport system can include up to three distinct mine wall layers. However, the finite-difference code employs zero-radon-concentration boundary conditions on the exterior mine wall surfaces, and is, therefore, not directly applicable to uranium mill tailings modeling and analysis.

3.3.5 Utah State University radon diffusion code

A FORTRAN code entitled FEREDA has been developed and published by Utah State University (Ph82) to model one-dimensional, steady-radon diffusion through porous, multilayer media. The solution methods and boundary conditions are equilvalent to the RAECO code developed by Rogers and Associates in 1980, but includes additional options for estimating values of the radon diffusion coefficients.

3.3.6 Other computational models and codes

A variety of other computational models and related computer codes (Cl74, Co181, Ed72) have been developed which are applicable to radon flux analysis and estimation for mill tailings systems. In general, these codes lack adequate documentation or are proprietary and thus not available for general use in the UMTRA Project.

4.0 COMPARISONS OF RADON FLUX MEASUREMENTS

Comparisons of radon flux measurement techniques have been performed previously, but they have usually been a quality-assurance check in connection with a larger research project. Several of these intercomparisons in the UMTRA Project have been reported recently (Ni81a, Har83) and have demonstrated equivalence of results from two or more methods being used in UMTRA research. However, data recently collected by the Technical Measurements Center (TMC) indicated that very large differences may exist in the results obtained by different methods used by different institutions (Kre82, No82). Although some of these differences are explainable by the theoretical considerations in Section 2.0, an experimental comparison of results from a common flux source was deemed necessary to compare the methods.

4.1 THIN-SOURCE REFERENCE RADON FLUX FACILITY

The TMC has constructed a thin-layer reference radon flux facility based upon a conceptual design by RAE. The thin-source flux facility avoids many of the temporal variations and the spatial variations typical of most tailings piles. Thus, it is useful for radon flux measurement comparisons. It can be shown from the radon flux equation for a bare tailings source that the thin configuration reduces uncertainty and time variations in the resulting radon fluxes. The one-dimensional flux equation for a bare tailings source is given as Equation 20. If the tailings layer is thin, however, Equation 20 reduces to:

$$J = 10^4 RE\rho\lambda x , \qquad (22)$$

which indicates the independence of the radon flux from the diffusion coefficient of the tailings. Since temporal flux variations are predominantly a result of changes in diffusion coefficient, the thin source should be more constant with time as well as more predictable from simple radium, emanation, density, and depth measurements.

The reference-source radon flux facility was prepared from tailings obtained from the Monticello, Utah tailings pile. The tailings were dried and thoroughly blended, and then spread in a 5.5-cm layer in a 1.8-m diameter metal tank. From twenty-five subsamples of this source material, the TMC estimates of the source parameters were $R = 883 \pm 35$ pCi/g, $E = 0.210$, $\rho = 1.51$ g/cm^3 and $x = 5.5$ cm. The radon flux estimated from these parameters was 32.3 pCi m^{-2}s^{-1}. Measurements completed in 1983 at the TMC on samples from the reference flux facility suggest a lower flux, however, with radium contents in the range of $R = 820 - 840$ pCi/g, and emanation coefficients of about $E = 0.21$. This suggests the reference flux may be on the order of 30.4 pCi m^{-2}s^{-1}.

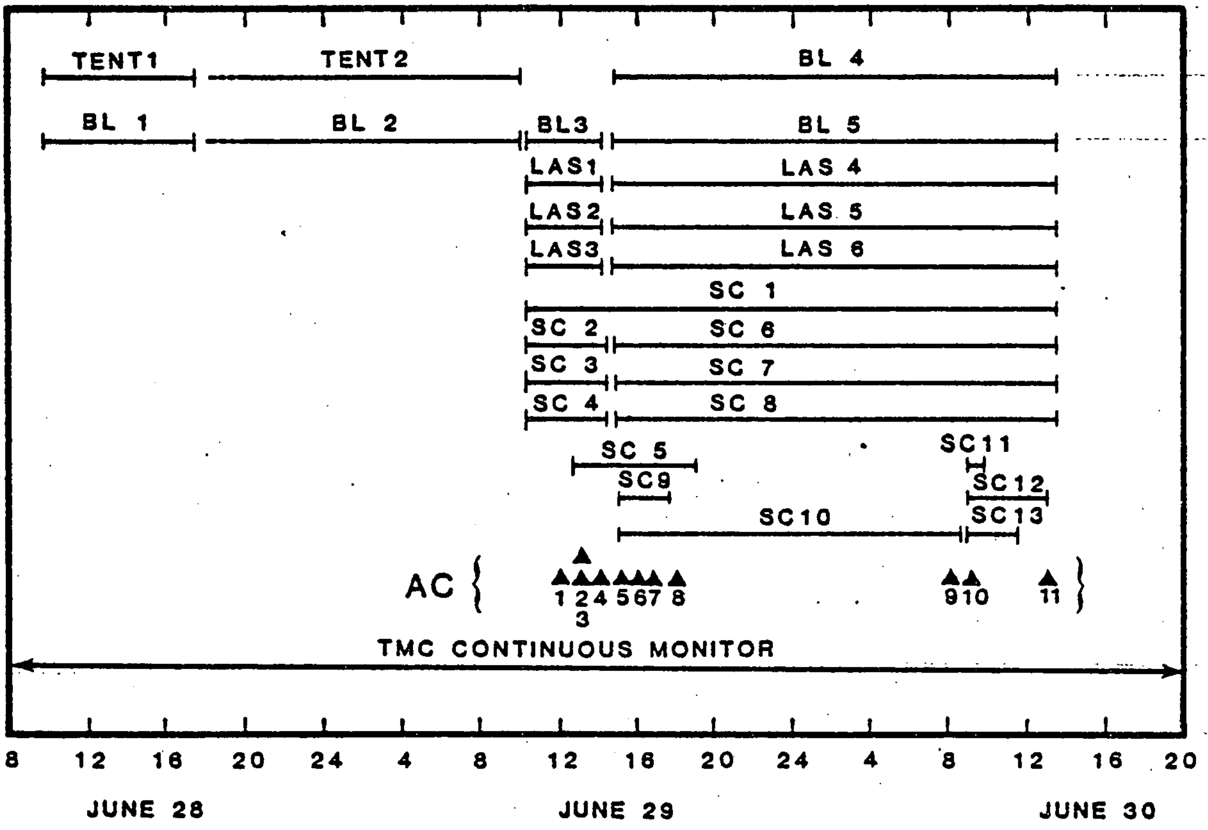
From previous experience in measuring emanation coefficients, a relative uncertainty of 5 percent is typically associated with emanation coefficients of this magnitude and tailings of this radium content. From the present radium measurements, it appears that an additional 2.4 percent uncertainty is associated with the radium measurements. Although the density can be accurately estimated from the total mass and volume of tailings utilized, the thickness of the tailings is constantly varying due to resmoothing after sample collection. Thickness measurements are made beneath each sampling location to normalize the measurement; however, these measurements are probably uncertain by about 2 mm, or 3.6 percent. The total uncertainty from these components is estimated to be about 7 percent; thus, the radon flux from the facility has an uncertainty of about 7 percent.

4.2 COMPARATIVE RADON FLUX MEASUREMENT PROCEDURES

It is of interest to compare numerous flux measurement techniques using the thin flux reference source. However, primary emphasis is placed on the techniques which were used extensively in the UMTRA research program and which thus provide a basis for current tailings cover design programs. Five principal methods were therefore compared on a preliminary basis to evaluate the technical aspects of these sampling and measurement methods. These methods included the accumulator-can method, the small charcoal canister method, two large-area charcoal adsorption methods, and the circulating-air, charcoal tent method. The comparisons are considered preliminary in several respects. Only a small number of samples could be collected with some of the samplers due to time, logistic and funding constraints. In addition, all samples were analyzed by one laboratory (RAE), with independent counting data available on only two of the samples. Although the flux measurement and associated analysis of all samples by a single laboratory is necessary in order to have consistent, accurate comparison of instruments and methods, this preliminary comparison of techniques is thus outside the traditional scope of an "interlaboratory comparison".

Sample collection was accomplished in two phases. In the first phase, Pacific Northwest Laboratory (PNL) personnel collected two sequential samples with the 0.929-m^2 charcoal tent sampler (Fre81) with simultaneous samples collected with a 0.107-m^2 "Big Louie" charcoal sampler. In the second phase, Rogers and Associates (RAE) personnel collected eleven samples with accumulator cans, thirteen with small charcoal canisters (0.0053-m^2), six with large charcoal samplers (0.16-m^2), and three with the Big Louie charcoal samplers. In addition, charcoal from one of the PNL tent and Big Louie samples was counted at the RAE facilities for comparison with PNL-supplied counting data. Figure 4.1 illustrates the relative time sequences for all of the sample collection. The Technical Measurements Center also operated a continuous flux monitor throughout the entire period to monitor possible temporal variations in radon flux. The physical arrangement of the samplers for the Phase II measurements is illustrated in Figure 4.2, which also shows the location of the tent sampler and the TMC continuous flux monitor.

The procedure for sample collection with the Big Louie samplers (Phase II) entailed loading of 400 g of activated charcoal into the 37-cm PVC sampler, spreading it to attain visual uniformity, replacing the horsehair spacer mat, and inverting the sampler onto the tailings surface. The sampler was then pressed about 1 cm into the tailings surface. After the sample collection period, the sampler was removed, and the charcoal was poured into metal cans and sealed for transport to the laboratory and gamma-ray analysis. The tailings



RAE-100770

FIGURE 4.1. RADON FLUX SAMPLE COLLECTION SEQUENCES FOR TENT, "BIG LOUIE" (BL), LARGE AREA CHARCOAL (LAS), SMALL CHARCOAL (SC), AND ACCUMULATOR CAN (AC) SAMPLERS.

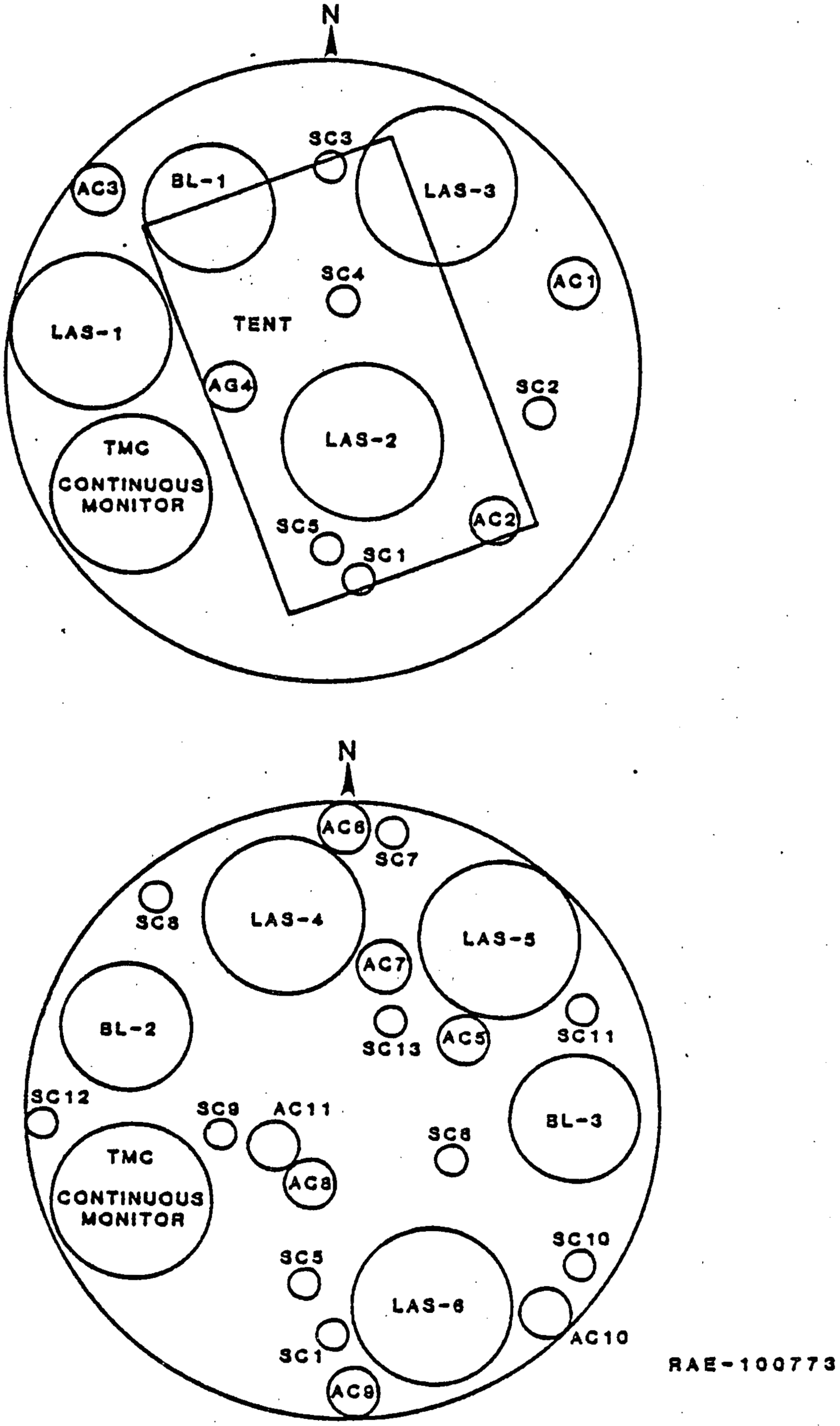


FIGURE 4.2. LOCATIONS OF INDIVIDUAL SAMPLES ON THE 1.8 m. DIAMETER THIN REFERENCE RADON FLUX SOURCE.

depth beneath the sampler was estimated by inserting a clean thin rod vertically into the tailings, withdrawing it, and then measuring the length of the dusty region.

A similar sampling procedure was followed with the large-area samplers (LAS). In this case, approximately 1000 g of activated charcoal was contained in a permeable nylon bag which was spread and smoothed over the surface to be sampled. A 46-cm metal cover was immediately placed over the bag and pressed 1 cm into the tailings. After the sample collection period, the cover was removed, and the bag was cleaned and sealed in a 15-cm diameter x 13-cm metal can for transport and gamma-ray analysis. The tailings depth beneath the sampler was measured as before.

The procedure with the small charcoal sampler was also similar, except in this case the charcoal consisted of a commercial type GMA gas mask canister (Mine Safety Appliance Co.). The canister was simply inserted into a metal can (8.3 cm dia x 4.5 cm) and covered with a Whatman #41 paper filter to keep the cartridge clean. The can was then inverted and pressed into the tailings for the desired sampling period. Upon removal, the tailings were cleaned from the surfaces, and the canister was sealed in its metal can. The tailings depth was also measured for the flux normalization.

For accumulator can samples a Lucas cell was evacuated and attached to the accumulator can (0.017 cm^2 x 26 cm) via a glass-fiber filter holder assembly. The accumulator can was pressed 1-cm into the tailings surface and allowed to remain for 5-16 min. The valve on the Lucas cell was then opened for sample collection, and then reclosed. The can assembly was then removed from the source and the tailings depth was measured. After a 4-h delay, the Lucas cell was counted using a phototube and scaler assembly, and it was then re-evacuated.

The tent and Big Louie samplers (Tent1 and BL1) were operated by PNL personnel as described in previous reports (Fre81, Har83). After sample collection and analysis, the charcoal samples were furnished to RAE, where they were sealed in metal cans for a different counting geometry and re-analyzed. This permitted a consistent basis for comparison of all charcoal sampling devices, and also allowed a comparison of the PNL and RAE gamma assay calibrations. The PNL calibration was based on an NBS radium standard (SRM-495-3C), and the RAE calibration was based on a DOE pitchblende ore standard (Si75). Both gamma-ray assays were performed with large lead-shielded NaI (Tl) scintillation detectors.

4.3 RADON FLUX MEASUREMENTS RESULTS

The results of the radon flux measurements described by Figures 4.1 and 4.2 are summarized in Table 4.1. As indicated, the means for each method ranged from 29.5 to 35.6 pCi $m^{-2}s^{-1}$, with an overall mean of the five methods being 31.5 ± 2.6. This is in excellent agreement with the value of 30.4 ± 2.1 predicted from radium and emanation measurements. The consistency of all of the five methods to accurately represent the radon flux from the thin source is also noteworthy, since three very different approaches are being compared. Excellent

TABLE 4.1 Radon fluxes measured by the various samplers,[a] normalized to 5.5 cm tailings thickness (pCi $m^{-2}s^{-1}$)

	SC	AC	LAS	Tent	BL
1	35.6	29.5	33.3	29.5[b,c]	28.8[d]
2	29.5	34.2	33.5		
3	32.5	31.2	26.9		41.6
4	32.9	27.1	29.7		31.9
5	37.0	29.8	28.2		33.3
6	30.2	22.4[f]	29.4		
7	28.2	31.4			
8	29.8	25.9			
9	32.5	18.3[g]			
10	30.2	33.4			
11	25.3[e]	23.7			
12	36.3				
13	34.6				
Mean	32.4	29.6	30.2	29.5	35.6
Std. Dev.	2.9	3.5	2.7	--	5.2
n	12	9	6	1	3

---

[a] SC = small charcoal sampler (0.0053 m^2); AC = accumulator can sampler (0.017 m^2); LAS - Large Area Charcoal sampler (0.164 m^2); Tent Sampler (0.929 m^2); and BL = "Big Louie" charcoal sampler (0.107 m^2).

[b] Flux based on the PNL count was 28.6 pCi $m^{-2}s^{-1}$.

[c] Other radon flux measurements using the tent with PNL counts were 29.6, 18.2, 29.2, and 35.9 pCi $m^{-2}s^{-1}$, giving a mean and S.D. for the PNL measurements of 28.3 ± 6.4 pCi $m^{-2}s^{-1}$.

[d] Flux based on the PNL count was 29.6 pCi $m^{-2}s^{-1}$. This measurement excluded from the mean because tailings thickness for normalization was unavailable.

[e] Excluded from mean because of short sample time and significant difference from remaining population.

[f] Excluded from mean because of excessive sample time and significant difference from remaining population.

[g] Excluded from mean because of leak in Lucas Cell observed in post-count evacuation.

agreement is also noted between the gamma assay calibrations of RAE and PNL in footnotes b and c of Table 4.1.

Since varied sample times were utilized in many of the small charcoal (SC) samples and the accumulator-can samples, the effects of extreme sampling time were examined in these data sets. Figure 4.3 illustrates the small charcoal (SC) flux results when plotted as a function of the sample collection times illustrated in Figure 4.1. As indicated, only the 1-h sample collection time gave an anomylously low result. When compared with the other SC flux data using a t-test, this point was found to be significantly different at the 95 percent confidence level, and was thus rejected in computing the mean in Table 4.1.

Figure 4.4 illustrates a similar plot of the radon fluxes measured with accumulator cans (AC) with varying sample collection times. The point designated by a triangle resulted from use of a leaky Lucas Cell, as discovered by the failure of this cell to maintain a vacuum after being counted. The 16-min sample was also rejected in computing the mean due to its exceeding the sample-time criterion and being significantly lower than the remaining flux measurements in the AC group.

An interesting comparison can be made of the present AC results with those from the other methods. As indicated in an earlier report (Ni81a), a 5 percent negative bias may be expected when 10-min sampling times are used with the present can geometry. Applying this 5 percent correction to the mean of 29.6 pCi $m^{-2}s^{-1}$ given in Table 4.1 gives a value of 31.1 pCi m^{-2} s^{-1}, which is in slightly better agreement with the charcoal sampling results. This correction has not usually been applied, however, since Lucas-cell efficiency factors (which are typically about 73 percent) vary by this much and are often not known with less than several percent uncertainty. An additional cause of negative bias in the AC measurements is the gradual loss of Lucas-cell efficiency with time. The Lucas cells used in this study have been used for several years, and have accumulated small scratches and deposits of vacuum grease on the optical windows, both of which would cause reduced efficiency for alpha-particle detection.

Application of the maximum sampling time criteria from Section 2.0 to the present sampling systems provides an interesting insight into the use of the systems. For the accumulator can samplers, the maximum sampling time is suggested to be on the order of 12.9 min, assuming an effective bulk diffusion coefficinet of D_e = 0.02 cm^2/s. For the small charcoal samplers, the air volume was estimated to be about 100 cm^3 and the charcoal mass about 40 g. From Equation 7, at 30°C, the radon distribution coefficient is K = 2973 pCi/g per pCi/cm^3. Then, from Equation 12, the maximum sampling time is estimated to be about 31 hours. As with the AC samples, however, the errors from exceeding this time criterion may still be small until it has been exceeded by a factor of two or more.

Similar analyses with the LAS samplers are based on 1000 g charcoal, and an air volume of 6200 cm^3, and give a maximum sampling time of 25 hours. For the BL samplers, a charcoal mass of 400 g and an air volume of 9400 cm^3 gives a maximum sampling time of 16 hours. For the tent sampler, the 400 g charcoal

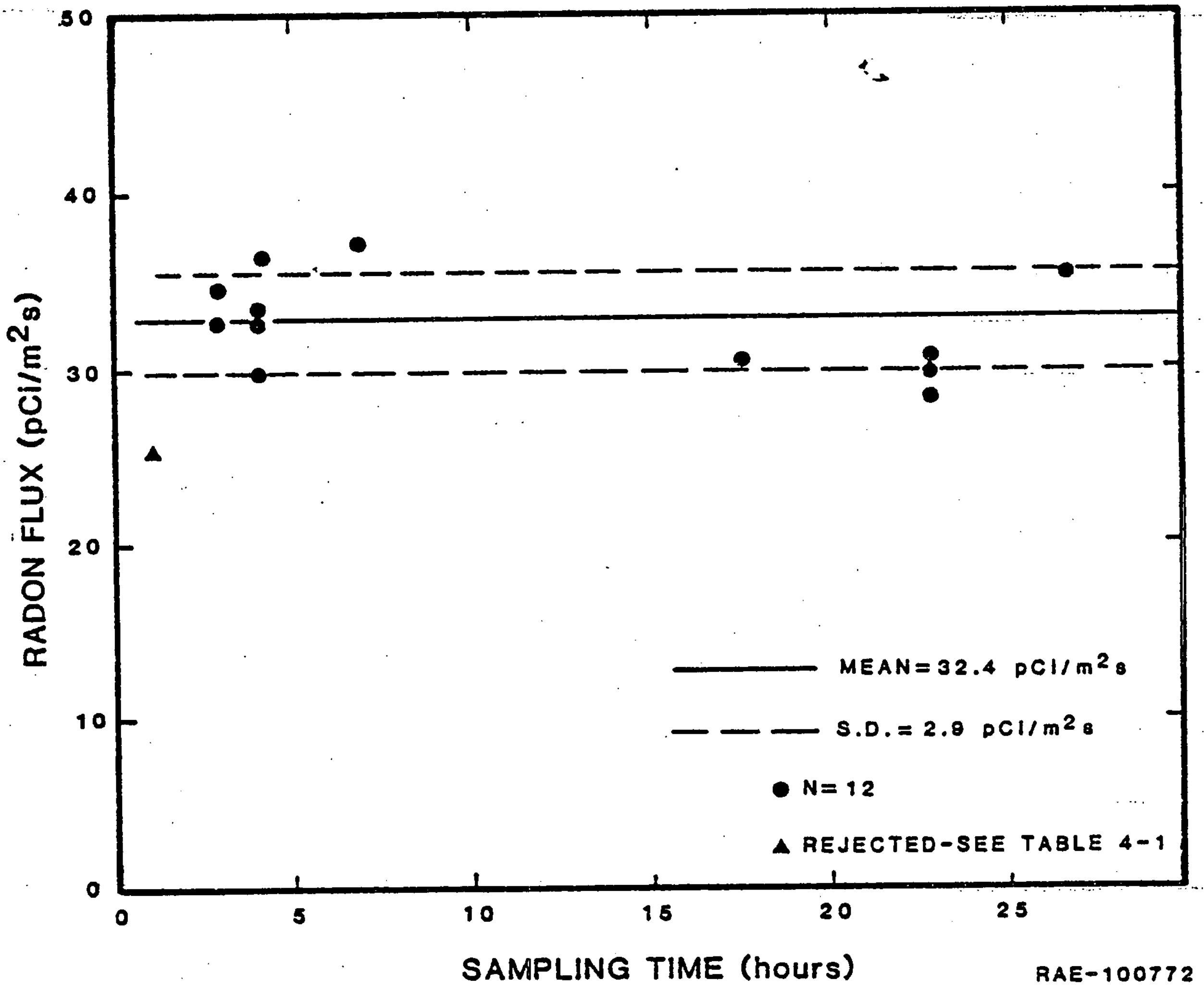


FIGURE 4.3. MEASURED RADON FLUXES VERSUS SAMPLING TIME FOR SMALL CHARCOAL (SC) SAMPLES.

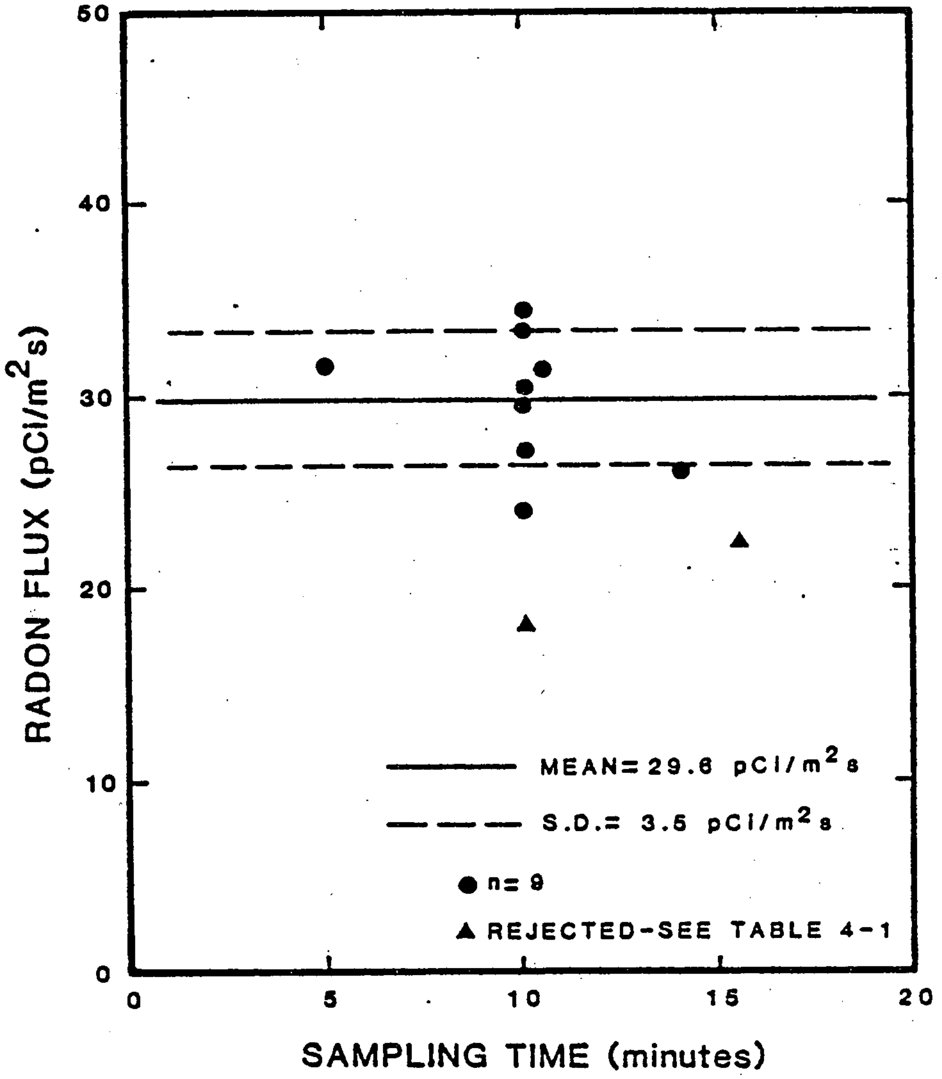


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FIGURE 4.4. MEASURED RADON FLUXES VERSUS SAMPLING TIME FOR ACCUMULATOR CAN (AC) SAMPLES.

mass and the 47,200 cm^3 volume give an approximate maximum sampling time of 1.8 hours. Although the sampling times of some of the charcoal samples exceeded the calculated maximum, all were within a factor of 2 of this limit except the tent samples, which showed more bias than the rest. Analysis of Equation 12 suggests that a greater ratio of charcoal mass to area sampled may help avoid this small bias.

In summary of the radon flux comparison measurements, it appears that the primary methods used in the UMTRA Program for radon flux measurement are all very well suited to give accurate results. Although precisions on the order of 9-15 percent were observed in this comparison, considerably greater uncertainties will be observed in field data and comparisons. These increased variations will be primarily a result of varying environmental parameters. The agreement among the various methods within about 10 percent suggests that they are capable of accuracies on the same order with proper calibration and careful procedures. Excessive sampling time appears to be the most likely source of bias with these methods, and should be evaluated before major applications by generating data as in Figures 4.3 and 4.4 for the expected sample-time range.

5.0 SUMMARY

The ability to accurately and precisely characterize fluxes of radon gas coming from soil surfaces is important in both regulatory and design aspects of remedial action for uranium mill tailings. The five primary methods used to measure radon fluxes in the UMTRA technology development research program range from simple passive charcoal adsorption samplers to a large, pumped-air tent system which also uses charcoal as a radon adsorption medium. Radon fluxes are measured by gamma-ray counting of the radon daughter activity on the charcoal, and dividing by the sampling area and time. A non-adsorptive technique utilizing a simple accumulator can with direct alpha-counting of the radon in the air is also described. The passive, radon-adsorption sampling methods can generally provide for integrated samples over one-day time periods, while the accumulator-can method is suitable for instantaneous (5 to 10-minute) grab samples. Maximum sampling times can be estimated for the accumulator can from its height, insertion depth, and the diffusion coefficient of the substrate. Maximum sampling times for the charcoal-adsorption samples depend primarily on the mass of charcoal, the area sampled, the insertion depth, and the diffusion coefficient of the substrate. Although large sample collection areas give improved precision, the improvement may be small compared to the area ratios and the extra effort associated with using the larger instruments. A few additional samples may actually provide for equivalent results between large and small collection devices, with considerable cost savings from the small samples. However, more data are needed to define this relationship.

A variety of mathematical methods are publicly available for computing radon transport calculations. For most radon flux predictions and tailings cover design problems, the one-dimensional, steady-state methods are adequate and are least expensive to perform. For analyzing special geometric conditions such as sampler performance, two- or three-dimensional numerical models are also available. Time-dependent, one-dimensional codes have also been developed, and have had primary application in simulating the effects of changing moisture conditions on radon fluxes. Three-dimensional, time-dependent capabilities are also available but have had little application in the UMTRAP research. Radon transport by both diffusive and diffusive-plus-advective processes can be handled by most of the mathematical methods; however, reliable data on advective velocities are seldom available to include in the calculations, nor are they usually of great long-term importance due to their self-canceling, cyclic nature.

An experimental comparison was made of the five principal flux measurement methods used in UMTRA research: the accumulator can, the small charcoal sampler, the large-area charcoal sampler, the "Big Louie" sampler, and the charcoal tent sampler. The comparison was conducted using the TMC thin-layer reference radon flux source, and thus avoided most time and space variations associated with previous comparisons. Excellent precisions and accuracies were demonstrated by all five of the methods, in contrast to the results of some earlier comparisons of other methods. Precisions expressed as relative standard deviations ranged from 8.9 percent for the large-area sampler to 14.6 percent for the "Big Louie". The accuracies were evaluated by comparison with the theoretical flux of 30.4 ± 2.1 pCi $m^{-2}s^{-1}$ predicted from the radium, emanation, depth and density measurements on the source. The respective errors of the five methods from this value were only -2.6 percent, 6.6 percent, -0.7 percent, 17.1 percent, and -3.0 percent with an average error of only 6.0 percent and an average bias of only + 3.5 percent. The average of the means of the five methods was 31.5 ± 2.6 pCi $m^{-2}s^{-1}$. Although slight effects were observed from excessive sampling times, these were small and were generally predictable.

It appears that present radon flux measurement technology is suitable for precise and accurate flux measurements at the levels involved in this study. Although the variations observed in this study were very small, much larger variations will usually be observed in field studies due to more variable radon source and diffusion conditions in both the time and space domains. The principles of operation of these five sampling methods are thus not dominant sources of error under most field conditions, as long as proper procedures and properly designed instruments are used. The most likely source of bias may often be excessive sampling time, which leads to low results due to radial diffusion around the sampler. Elevated temperatures decrease the maximum sampling times for charcoal samplers, and experimental tests of sampling-time effects should usually be conducted under the expected sampling conditions if sampling times approach or exceed the maximum values.

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Durango, Colorado, DOE/UMT 103, FBDU-360-06
Slick Rock, Colorado, DOE/UMT-0115, FBDU-360-07
Naturita, Colorado, DOE/UMT-012, FBDU-360-08
Grand Junction, Colorado, DOE/UMT 105, FBDU-360-09
Rifle, Colorado, DOE/UMT 108, FBDU-360-10
Maybell, Colorado, DOE/UMT 116, FBDU-360-11
Gunnison, Colorado, DOE/UMT 107, FBDU-360-12
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Green River, Utah, DOE/UMT-0114, FBDU-360-14
Converse County, Wyoming, DOE/UMT-0119, FBDU-360-15
Falls City, Texas, DOE/UMT 111, FBDU-360-16
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